# Trace-element modelling of mare basalt parental melts: Implications for a heterogeneous lunar mantle

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## ABSTRACT

The heterogeneous-source model of mare basalt formation indicates that Lunar Magma Ocean (LMO) overturn produced an uneven mixture of early olivine-pyroxene, and late-stage, ilmenite-rich cumulates, which subsequently partially re-melted to give rise to mare magmas. These heterogeneous cumulate source regions would not only have been characterised by different mineral modal abundances, but also by different trace element compositions. The aim of this work was to investigate the petrology and geochemistry of a diverse suite of Apollo mare basalts, and utilise trace-element modelling in order to understand their petrogenetic history. Chemical modelling confirms the mare basalts were produced by small degrees of LMO cumulate partial melting (<10 %), and that the dominant melting type (batch vs. fractional) changes between different basalt groups. Similarly, a single source mineralogy cannot be applied to all mare basalt types, implying the lunar mantle was laterally heterogeneous at the time of mare basalt melt production. Plagioclase is not required in the source of most mare basalts, with the notable exception of the Apollo 14 high-Al basalts. Addition of more than 1 % plagioclase to the source of other basalts produces weaker negative Eu anomalies than those observed in the samples. AFC calculations emphasize the compositional differences between the assimilated material incorporating into the Apollo 14 high-Al and Apollo 11 high-K mare basalt partial melts, highlighting the complexities of mare basalt petrogenesis.

## 1. INTRODUCTION

Shortly after the return of first lunar samples to the Earth, initial investigations identified pure anorthosite grains in Apollo 11 samples. This observation led to the development of a hypothesis that a global-scale compositional separation (differentiation) had occurred on the Moon after its formation (Smith et al., 1970; Wood et al., 1970a,b). This differentiation is thought to be the result of a large scale global melting, which produced a lunar magma ocean (LMO) (Smith et al., 1970; Wood et al., 1970a,b; Warren and Wasson, 1979a,b; Warren, 1985; Wieczorek and Zuber, 2001). It is proposed that the LMO could only have been created by the high pressures and temperatures produced during lunar accretion (Pritchard and Stevenson, 2000). Estimates of magma ocean depth vary from 200 km to whole Moon melting (e.g., Warren, 1985; Ryder, 1991; Neal, 2001; Elkins-Tanton et al., 2002). These estimates are based on mass balance arguments, taking into consideration the abundance of plagioclase and incompatible elements in the crust, in addition to estimates of the depth of mare basalt source regions. Based on Hf-W and Sm-Nd isotope data on lunar samples, it has been proposed that the LMO crystallised 30-50 Myr after the start of the Solar System (e.g., Kleine et al., 2002). However, more recent data implies that the LMO crystallisation was protracted - the last remnants of the LMO crystallised after $^{182}$Hf became extinct at approximately 60 Ma (Touboul et al., 2007; Borg et al., 2011). In addition, recent $^{146}$Sm-$^{142}$Nd isotopic data indicate that the mare basalt source regions were closed with respect to these isotopic systems around 150–190 Ma after the accretion of the Moon (Brandon et al. 2009).

The current models of the LMO crystallisation include the formation of early-forming olivine and low-Ca pyroxene crystals (e.g., Snyder et al., 1992a; Elardo et al., 2011), which

would have had a higher density than the surrounding melt, and so sank towards the bottom of the LMO, producing cumulate layers. The anorthitic plagioclase feldspar was the next liquidus phase with a lower density relative to the residual melt, and, thus, it floated towards the surface of the LMO to form an anorthositic primary crust (e.g., Smith et al., 1970: Wood et al., 1970a,b). After > 95 % LMO crystallisation, significant amounts of ilmenite also crystallised at ~ 100 km depth (Warren, 1985; Ryder, 1991; Shearer and Papike, 1999). This depth estimate is based on the fact that ilmenite crystallisation requires at least 4 wt % $TiO_2$ content in the melt (Hess et al., 1978), and crystallisation of a magma ocean cannot produce such high $TiO_2$ at depths below ~100 km, unless the bulk Moon has exceptionally high $TiO_2$ (>1 wt %, Ryder, 1991). Ringwood and Kesson (1976) argued that the presence of very high-density ilmenite-rich cumulates above less dense olivine and pyroxene cumulates caused a density instability, resulting in mantle overturn and the sinking of ilmenite-rich cumulates, possibly to the core-mantle boundary (Hess and Parmentier, 1995; Zhong et al., 2000; de Vries et al., 2010). It has been hypothesised that the sunken high-Ti cumulates subsequently partially re-melted and contributed to the formation of high-Ti mare basalts and picritic glasses (Snyder et al., 1992a; Hess and Parmentier, 1995; Zhong et al., 2000). There has been some debate as to whether or not high-Ti melts are buoyant below 200-300 km on the Moon (Circone and Agee, 1996; Wagner and Grove, 1997; van Kan Parker et al., 2012), but the multiple saturation depths of the high- and low-Ti picritic glasses (which, provided the melt is primary, indicate the depth and temperature of a melt's segregation from its source region) are all below this depth, at > 250 km (Green et al., 1978; Delano, 1980; Wagner and Grove, 1997; Van Parker et al., 2012). In addition, these multiple saturation points indicate that none of the picritic glasses have ilmenite on the liquidus at any pressure, suggesting that either the melt fraction was high enough to exhaust the high-Ti phase in the source, or that Ti was assimilated after melting (Elkins-Tanton et al., 2002 and references

100 therein). The multiple saturation depths of the high-Ti mare basalts are all above 300 km.
101 Therefore, the high-Ti picritic glass melts could have assimilated Ti from the Ti-rich
102 cumulates above their source regions, in the region of mare basalt melt production (Elkins et
103 al., 2000; Elkins-Tanton et al., 2002).
104
105 A slightly different view of LMO crystallisation emerges if garnet is assumed to be present
106 in the lunar mantle, as suggested by Neal (2001). A garnet bearing lunar mantle would imply
107 a bulk Moon with a higher $Al_2O_3$ content than in the models discussed above. If this is the
108 case, and either the LMO was deep (possibly whole Moon) or the deep interior didn't melt,
109 plagioclase may have crystallized earlier than other models predict. This results in early
110 cumulates having an inherent negative Eu anomaly and may negate extensive cumulate
111 overturn (in agreement with the model of Snyder et al., 1992a).
112
113 Regardless of the exact mechanism, the process of lunar mantle overturn/mixing is thought
114 to have resulted in chemical/mineralogical heterogeneity in the cumulate lunar mantle (e.g.,
115 Binder, 1985; Hughes et al., 1990; Van Orman and Grove, 2000; Neal, 2001; Elkins-Tanton
116 et al., 2002; Shearer et al., 2006 and references therein; Giguere et al., 2010; Elkins-Tanton
117 et al., 2011). This mare basalt source region heterogeneity is evident from wide
118 compositional and isotopic variations in basalts originating from similar crustal depths (e.g.,
119 Rhodes et al., 1977; Beaty and Albee, 1978; Baldridge et al., 1979: Neal et al., 1990a,b; Neal
120 and Taylor, 1992; Beard et al., 1998; Neal and Kramer, 2006; Rankenburg et al., 2006). An
121 example of this heterogeneity is found at the Apollo 12 site, where three mare basalt groups
122 were identified (olivine-, pigeonite- and ilmenite-rich) that appear to be derived from
123 separate source regions based on their Nd isotopic data (e.g., Snyder et al., 2000). In

addition, the lunar meteorites provide further evidence for Nd isotope variations in source-region compositions (e.g., Borg et al., 2009).

In order to further improve our understanding of the compositional characteristics of the mare basalt source regions, and their derivative basaltic partial melts, we conducted major and trace element analyses of a compositionally diverse suite of mare basalt samples from the Apollo collection. A number of these samples have not been studied extensively in the past (or at all in the case of some trace elements [SOM appendix 1]), and others have not been analysed with modern instrumentation and techniques. Therefore, our data represent the most comprehensive geochemical dataset on these samples. Subsequent to these analyses, geochemical modelling was carried out to determine the degree of partial melting, crystal fractionation, and assimilation to account for different mare basalt compositions.

## 2. MARE BASALT CLASSIFICATION AND THE SAMPLE SET

From mare basalt bulk-rock compositions it became apparent that the most useful lithologic discriminant is bulk-rock $TiO_2$ content. Papike et al. (1976) and Papike and Vanimann (1978) defined three compositional groups based on bulk-rock $TiO_2$ content in mare basalts: very low-Ti basalts (< 1 wt % $TiO_2$), low-Ti basalts (1-5 wt % $TiO_2$) and high Ti-basalts (9-14 wt % $TiO_2$). Neal and Taylor (1992) argued that secondary and tertiary divisions can be made using $Al_2O_3$ (i.e., < 11 wt % = low-Al, > 11 wt % = high-Al) and K contents (i.e., < 2000 ppm = low-K and > 2000 ppm = high-K). These divisions yield a classification containing twelve possible categories, of which six have been identified in the Apollo and Luna collections (Fig. 1). Of these six groups, five groups (22 samples) are represented in the

149 sample set we have analysed (Table 1), the very low-Ti, low-Al, low-K basalts are not
150 represented.
151
152 We also analysed one KREEP basalt (sample 15386). Isotopic studies show KREEP basalts
153 erupted prior to the majority of mare basalts (e.g., Nyquist et al., 1975; Carlson and Lugmair,
154 1979), and are enriched in incompatible elements including potassium, rare earth elements
155 (REE) and phosphorus. Incompatible element enrichment indicates that the KREEP basalt
156 source material (urKREEP) was a residual melt, which formed after > 99 % crystallisation of
157 the LMO (e.g., Warren and Wasson, 1979a,b). However, more recent studies indicate that
158 KREEP basalt source material must have been highly magnesian, based on mineral and
159 bulk-rock chemical data (Rankenburg et al., 2006; Taylor et al., 2012). It is, therefore,
160 probable that urKREEP residual material was mixed with Mg-rich orthopyroxene and
161 olivine cumulates (which crystallised early during LMO solidification), to produce the
162 KREEP basalts (for further details see Rankenburg et al. 2006; Taylor et al., 2012; Elardo et
163 al., 2012).
164
165
166 **3. ANALYTICAL TECHNIQUES**
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168 We were allocated 22 polished thin sections of mare basalts by the Curation and Analysis
169 Planning Team for Extraterrestrial Materials (CAPTEM). Back-scattered electron (BSE) and
170 X-ray images of each thin-section, along with mineral compositional data, were obtained by
171 Energy Dispersive Spectrometry using a JEOL 5900LV SEM at the Natural History Museum,
172 London (NHM). This instrument was fitted with an Oxford Instruments INCA energy
173 dispersive X-ray micro-analyser system, and operated at a 20 keV accelerating voltage and 2

nA beam current. Magnifications of ≥ 200 were used for whole thin-section images (depending on the sample-size and texture in each thin-section), producing a pixel size of 1-10 µm. The modal mineralogy of each sample was calculated using X-ray elemental image data and the Phasemap function of Oxford Instruments INCA software, which allowed identification of pixels representing each mineral phase (e.g., Joy et al. (2006)). Based on the standard deviation of five consecutive analyses of the same thin-section, the uncertainty related to this technique is ca. ± 1 vol %. This uncertainty can be further reduced for the detection of minor minerals by additional analyses of the X-ray elemental images using image processing software. This modal recombination technique is the most precise technique for obtaining two-dimensional bulk compositions, when used alongside mineral chemical data (e.g., Albee et al., 1977; Berlin et al., 2006). Major element composition of minerals were determined using a Cameca SX100 electron microprobe at the Natural History Museum, London, UK. The instrument was operated at a 20 KV accelerating voltage with a 20 nA beam current and a focused beam (5 µm for plagioclase and apatite, 1 µm for all other phases). We used 10–30 s count time per element, and employed well-characterized NHM standards (Na – jadeite, Mg - synthetic forsterite, Al – synthetic corundum, Si + Fe - synthetic fayalite, K – orthoclase, Ca – wollastonite, Ti + Mn - synthetic Mn-Ti oxide, Cr – synthetic Cr-oxide, Co + Ni – Co-Ni synthetic metal, P – synthetic Sc phosphate).

We were also allocated ~250 mg rock chips of the same 22 mare basalts, which were used for bulk rock geochemical analyses. These chips were converted into homogeneous powdered samples under clean laboratory conditions (better than class 1000) using two agate pestles and mortars at the Open University, UK. Before and after each sample was powdered the pestles and mortars were cleaned in turn with acetone, pure quartz sand, followed by acetone and isopropanol.

Bulk-rock concentrations of major and minor elements were determined using inductively coupled plasma atomic emission spectrometry (ICP-AES; Varian Vista Pro Axial) at the NHM. A 40 mg sub-split of each homogenised sample powder was pre-treated with 0.5 ml concentrated $HNO_3$ (Aristar® grade, VWR) and afterwards fused with 120 mg of $LiBO_2$ in a Pt/Au crucible. All forms of sulphur are lost during the fusion (ca. 1000 °C), therefore, bulk-rock sulphur content cannot be determined using this technique. The fused beads were dissolved in 40 ml 10% $HNO_3$ and made up to 100 ml with Milli-Q water. Calibration was performed using certified reference materials (CRM) prepared in the same way: BCR-2, BHVO-2 (basalts, USGS), DTS-1, DTS-2B (dunites, USGS), GSP-1 (granodiorite, USGS), JG-1 (granodiorite, GSJ), Mica-Fe (biotite, CRPG), NIM-G, NIM-L, NIM-P (granite, lujavrite and pyroxenite; NIM, South Africa) and SU-1 (nickel-copper-cobalt ore, CCRMP, Canada). Calibration curves were calculated using up to 15 points for each element. For elements present in some samples at concentrations above the calibration range (Ti, Ca, Fe), the solutions were further diluted (1:6 with $LiBO_2$ solution in 4% $HNO_3$) and re-analysed. Each sample (diluted and undiluted) was analysed in triplicate. Analytical precision for each major element was always better than 1 %, and in 75 % of cases it was better than 0.6 %. Accuracy was checked by the analysis of CRMs NIM-D (dunite, NIM) and PCC-1 (peridotite, USGS) and was within 1.5 % of recommended values for all elements present as majors (> 1 wt%) and within 9 % for all other elements. The instrument was recalibrated every 60 samples, and within a given calibration the instrument drift was monitored and corrected by analysing the BHVO-2 standard every 20 samples (the drift never exceeded 10 % of the value).

Bulk-rock trace element abundances were determined using an Agilient 7500a inductively coupled plasma mass-spectrometer (ICP-MS) at the Open University, UK. Oxide interferences were kept low at 0.3 % $CeO^+/Ce^+$ and doubly charged species at ≤ 1 % ($Ce^{++}/Ce^+$). A 0.01 g sub-split of each sample powder, along with the same amount of relevant reference materials and procedural blanks, were digested in pre-cleaned Teflon vials using a few drops of concentrated $HNO_3$ and 200 µl of concentrated HF. After 48 hours on a hotplate at 130 °C the solution was dried down, and refluxed in 1 ml 6M HCl for 24 hours, again at 130 °C. Following this the samples were refluxed in 1 ml conc. $HNO_3$ for a further 24 hours. Then the samples were taken up in 0.6 ml conc. $HNO_3$ and 0.6 ml 6M HCl, before being transferred into a pre-cleaned HDPE vial. These sample solutions were made up to 20 ml with ultra-pure water (with resistivity higher than 18.2MΩ). All acids used in the dissolution procedure were Teflon distilled (TD) grade. USGS certified reference materials (CRMs) andesite AGV-1, dolerite DNC-1, diabase W-2, and basalts BIR-1 and BHVO-1 were used for accuracy check (Eggins et al., 1997; see SOM Table S1). An internal standard solution, containing Be, Rh, In, Tm, Re and Bi, was added to all samples. The basaltic CRM DNC-1 was also analysed as an "unknown" after every 6 samples, together with a 2 % $HNO_3$/HCl blank, in order to monitor instrumental drift and to assess the external reproducibility of the data. The measured concentrations of elements in DNC-1 are routinely within 11 % difference of published values (Eggins et al., 1997), and commonly show < 5 % difference (SOM, Table S2). Exceptions include Rb, Nb, Cs, Ba, Th and U, all of which plot within 20 % difference of published values, except Cs which plots within 29 %. Detection limits for trace elements with atomic masses greater than 85 are usually less than 10 ppt in solution (equivalent to 20 ppb in the rock), but are somewhat higher for lighter elements (less than 100 ppt in solution, 200 ppb in the rock).

## 4. PETROGRAPHY AND BULK-ROCK DATA

The modal mineralogy of each sample is summarised in Table 2, and is in good agreement with previously published data (Meyer, 2011, and references therein). In terms of mineral abundances, the only systematic difference between the low- and high-Ti basalts appears to be their ilmenite content. The samples are dominated by clinopyroxene, commonly consisting of Mg-rich pigeonite/augite cores with Fe-rich rims (see Table S3 for mineral chemistry). Plagioclase and ilmenite are the next most abundant mineral phases, and textural evidence suggests that they began to crystallise after crystallisation of clinopyroxene cores in majority of the samples. Where olivine is present, it generally appears to have been one of the first phases to crystallise, along with Cr-spinel, rare armalcolite, and Mg-rich clinopyroxene cores (see also Papike et al, 1976). However, a number of samples (e.g., the Apollo 15 olivine normative basalt samples 15016 and 15555) contain olivine grains with Fe-rich rims indicating protracted olivine crystallisation in these samples. Accessory minerals include armalcolite, apatite, (REE)-merrilite, troilite, Cr-Ti spinel, Fe-Ni metal, silica, pyroxferroite, tranquillityite, zirconolite and baddeleyite (Table S3).

### 4.1. Major elements

Consistent with petrographic observations, bulk-rock $TiO_2$ data exhibit a dichotomy between low- and high-Ti samples, both for our sample set and all previously published data (Table 3, Fig. 1). One possible exception is Apollo 11 low-K basalt 10058,256, which lies just inside the intermediate-Ti region (8.66 wt % $TiO_2$). Sample 10058 has shown a $TiO_2$ wt % variation

in previous datasets, ranging between 9 and 10.6 wt % (LSPET, 1969; Duncan et al., 1976; Beaty and Albee, 1978; Jerde et al., 1994). This variation could be ascribed to the large size and possible heterogeneous distribution of ilmenite grains in sample 10058 (e.g., Simpson and Bowie 1970; Brown et al., 1970; Beat and Albee, 1978). The $Al_2O_3$ content of sample 15555,982 is the same as the highest previously reported value for this sample (Cuttita et al., 1973). Previously published data show a wide variation in 15555 $Al_2O_3$ (7.45-10.32 wt %) (Meyer, 2011, and references therein). These differences appear to be related to sample heterogeneity. 15555 was the second coarsest sample collected at the Apollo 15 site, with pyroxene grains up to 5 mm (Ryder, 2001). Hence, previously published plagioclase modal abundances vary from 25 to 35 vol % (Longhi et al., 1972; McGee et al. 1977; Heuer et al., 1972; Nord et al., 1973). All other major element abundances for these samples are consistent with previously published data (Fig. 1).

On a plot of $TiO_2$ vs. $Al_2O_3$ (Fig. 1a), the majority of Apollo 11 and 17 basalts in our sample set cluster together in the high-Ti, low-Al region. Apollo 11 low-K basalt 10058,256 is the exception, as it plots just within the high-Al region. This high-Al content is in agreement with previously published data for this sample (10.2-13.0 wt % $Al_2O_3$: LSPET, 1969; Duncan et al., 1976; Beaty and Albee, 1978; Jerde et al., 1994). In addition, previously published data indicate that a number of samples collected from the Apollo 11 and 17 sites were of high-Al and/or intermediate-Ti compositions (e.g., LSPET, 1969; Engel and Engel, 1970; Morrison et al., 1970; Turekian and Kharkar, 1970; Kharkar and Turekian, 1971; Wakita et al., 1970; Wänke et al., 1970; Engel et al., 1971; Smales et al., 1971; Laul et al., 1975; Warner et al., 1975; Duncan et al., 1976; Beaty and Albee, 1978; Ma et al., 1980; Jerde et al., 1994). The low-Ti basalts of Apollo 12 and 15 in our sample set show greater scatter in $Al_2O_3$ content than the high-Ti basalts, in accordance with previously published values for these basalt

298 groups (Fig. 1a). Our analytical data for ilmenite basalt 12064 indicate that this sample
299 contains ≥ 11 wt % $Al_2O_3$, which is in agreement with previously reported range of 10.39-
300 12.85 wt % $Al_2O_3$ (LSPET, 1970; Ganapathy, 1970; Scoon, 1971; Kushiro and Haramura,
301 1971; Warren et al., 1986; Waren et al., 1987). Apollo 15 KREEP basalt 15386 and Apollo
302 14 high-Al basalt 14053 also plot in the high-Al region, in agreement with previously
303 reported values for these samples (e.g., LSPET, 1971; Willis et al., 1972; Hubbard et al.,
304 1972; Rhodes and Hubbard, 1973; Warren and Wasson, 1978).
305
306 Compared with mare basalts, Apollo 15 KREEP basalt 15386 contains elevated $K_2O$ (0.64 wt
307 %), although previously published Apollo 15 KREEP data show $K_2O$ contents extending
308 downwards and overlapping with mare basalt abundances (Fig. 1b). The Apollo 11 high-K
309 basalts contain approximately half of this amount (0.34 to 0.31 wt %), and the Apollo 14 low-
310 Ti, high-Al basalt 14053 contains even lower $K_2O$ (0.12 wt %). However, previously
311 published data show that a number of Apollo 14 mare basalt samples do contain > 0.24 wt %
312 $K_2O$ (equivalent to 2000 ppm potassium) (e.g., Dickinson et al., 1985; Shervais et al., 1985a;
313 Neal et al., 1988b). These samples can, therefore, be classified as high-K using the
314 classification of Neal and Taylor (1992). The Apollo 14 data envelope shown in Fig. 1b does
315 not include the very high-K (VHK) Apollo 14 basalts, as this group is not represented in our
316 sample set – VHK samples contain between 0.35 and 1.86 wt % $K_2O$ (e.g., Duncan et al.,
317 1975; Shervais et al., 1985b; Neal et al., 1988b; Neal et al., 1989). All Apollo 12 and 15 low-
318 Ti basalts contain $K_2O$ below 0.24 wt % and are, therefore, classified as low-K.
319
320 The bulk-rock MgO content varies from 6.1 to 16.4 wt % in our sample set (Fig. 1c), where
321 only the low-Ti Apollo 15 basalts 15016 and 15555, Apollo 12 olivine basalt 12040, and
322 Apollo 12 ilmenite basalt 12016 contain > 10.5 wt % MgO, in accordance with previously

published data (e.g., Compston et al., 1971; Kushiro and Haramura, 1971; Scoon, 1971; LSPET, 1972; Chappell and Green, 1973; Rhodes et al., 1977; Neal et al., 1994a,b; Ryder and Schuraytz, 2001). The high MgO content of these four basalts appears to be related to olivine accumulation (see SOM appendix 1 and discussion section). The remaining Apollo 12 basalts show a relatively restricted variation in terms of MgO content (6.4-7.7 wt %), although previous data for other Apollo 12 basalts indicate a much wider variation in MgO content, from ~6-17 wt % (Fig. 1c). Apollo 14 high-Al basalt 14053 and Apollo 15 KREEP basalt 15386 show similar MgO contents (7.8 and 8.2 wt %, respectively), although previously published data indicate a number of Apollo 15 KREEP basalts containing lower MgO contents than any of the Apollo 14 high-Al basalts (Fig. 1c). The high-Ti basalts display more restricted compositions, with Apollo 17 basalts showing higher MgO (8.1-10.1 wt % for our sample set; 4.41-11.93 wt % for all previously analysed samples) than the Apollo 11 basalts (6.1-7.9 wt % for our sample set, 3-10 wt % for all previously analysed samples, Fig. 1c). Apollo 17 type A basalt 75055 shows the lowest MgO value of all the Apollo 17 samples in our sample set (8.1 wt %), and plots closest to Apollo 11 basalt 10020 (7.9 wt %). The limited variation in bulk-rock MgO content of the high-Ti mare basalts compared to the low-Ti basalts is consistent with the observations made in previous studies (Fig. 1c). Lunar basaltic meteorites (Asuka 881757, MIL 05035, LAP 02224, LAP 02205, Dho287, NEA 003, NWA 4898 and Y793169, see Fig. 1 for references) contain $Al_2O_3$, $K_2O$ and MgO at the level similar to the low-Ti basalts, but their $TiO_2$ content extends into the region defined by the very low Ti (VLT) mare basalts.

346 **4.2. Compatible trace elements**

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348 In all cases, the compatible element data of this study are consistent with previously
349 published values for these samples (Fig. 2), or samples from the same group where previous
350 data from the same sample are not available (e.g., vanadium contents in 12047 and 75055).
351 Decreasing V and Cr content with decreasing Mg# in the high-Ti mare basalts suggests
352 ilmenite and Cr-spinel fractionation in their parental magmas (V is highly compatible in
353 ilmenite). Scandium is most abundant in high-Ti basalts, but its abundance does not change
354 significantly with Mg# (Fig. 2a). Apollo 15 basalts (both KREEP and olivine-normative)
355 generally contain the lowest Sc concentrations and also show no variation with Mg#. Apollo
356 12 low-Ti basalts show increasing Sc concentrations with decreasing Mg#; the average
357 olivine and pigeonite basalt Sc abundances are slightly lower than those of the ilmenite
358 basalts, but there is a large degree of overlap between the two groups. Most of the measured
359 basaltic meteorites show a continuation of the Apollo 12 trend in this plot, but MIL 05035
360 and Asuka-881757 fit more closely to Apollo 11 values. Vanadium is highly compatible in
361 ilmenite, but V is more abundant in the low-Ti basalts than in the high-Ti basalts, which
362 contain more ilmenite (Fig. 2b). No significant variation in V abundance is apparent with
363 changing Mg# in the low-Ti basalts. The basaltic lunar meteorites plot between the two
364 groups. Apollo 17 type B2, C and unclassified basalts 70017 and 70035 contain slightly
365 higher V abundances than the type A and Apollo 11 basalts (see Table 1 for basalt type
366 characteristics – separate basalt types are thought to originate from distinct parental melts).
367 Both groups show a decrease in V concentration with decreasing Mg#, though the trend is not
368 as coherent as that observed for Sc contents in Apollo 12 samples. The same trend is visible
369 for Cr content in the Apollo 17 basalts in this instance along with the Apollo 12 basalts (Fig.
370 2c). The Apollo 15 basalts show no Cr vs. Mg# trend, and the Apollo 11 basalts show only a

371 weak positive correlation. Apollo 14 high-Al basalts show increasing Sc, V and Cr contents
372 with decreasing Mg#. The Cr content of the basaltic lunar meteorites varies more widely than
373 those of the Apollo samples, whereas Sc and V contents are more restricted (although this
374 may be due to the smaller meteorite sample set).
375
376
377 **4.3. Incompatible trace elements**
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379 Bulk-rock REE data for mare basalts in the Apollo sample collection have been previously
380 published by a number of researchers over the past forty years. As a result there exists an
381 extensive dataset, but in most cases only selected REEs were analysed in a given sample
382 because of the limitations posed by instrumentation and analytical techniques. This study
383 reports one of the most comprehensive and complete REE datasets currently available for the
384 mare basalts. Our data are in good agreement with previously published values (SOM
385 appendix 2), except for sample 15555,982, which does not seem representative of the bulk-
386 rock composition. 15555,982 contains systematically lower REE concentrations than all other
387 previous analyses of this sample, which indicate levels similar to sample 15016 for these
388 elements (0.14-4.94 vs. 0.25-14.9 ppm, see appendix 3). This is most likely because of the
389 large crystal sizes in sample 15555, which contains plagioclase crystals up to 3 mm long and
390 clinopyroxene crystals up to 2 mm long. An over representation of plagioclase would explain
391 the slightly positive europium anomaly in 15555,982, and the slight LREE enrichment
392 (appendix 3). Disparities among bulk-chemical abundances have been previously noted by
393 Neal (2001), Ryder and Schuraytz (2001), and Spicuzza et al. (2007). As the former two
394 studies determined bulk trace element abundances of a 4 g chip whereas the chip analysed for

this study was 250 mg, it is perhaps not surprising that our REE measurements do not reflect bulk-rock abundances for this exceptionally coarse-grained sample.

Apart from sample 15555, Apollo 15 low-Ti basalt 15016 has the lowest REE abundances of all the samples analysed in this study, along with Apollo 12 olivine basalt 12040 (12-23 times chondrite normalised (CN) values, when normalised to the CI chondrite data of Anders and Grevesse (1989) (Fig. 3). The remaining Apollo 12 samples display REE abundances varying from 17 to 43 times CN values. All these low-Ti samples show relatively flat trends and weakly negative europium anomalies (Eu* = 0.56-0.65; where Eu* = chondrite-normalised $Eu/\sqrt{(Sm \times Gd)}$; value > 1 is indicative of a positive anomaly and a value < 1 is indicative of a negative anomaly). In our sample set the high-Ti basalts of Apollo 17 have higher HREE abundances than the low-Ti basalts of Apollo 12 and 15 (41-93 vs. 13-41 times CN values, respectively). However, LREE depletion within the Apollo 17 basalts (15-86 times CN values) results in some overlap with the LREE contents of the Apollo 12 and 15 samples (15-41 times CN values). Apollo 11 low-K basalts contain lower REE abundances than the high-K basalts collected at the same site, and show a slightly larger spread in data (52-110 vs. 101-154 times CN values for the HREE). The former is depleted in LREE to a degree similar to the Apollo 17 basalts ($La/Sm_n$ = 0.66-0.85 and 0.58-0.76, respectively), whereas the latter have much flatter trends ($La/Sm_n$ = 1.27 for all four samples analysed in this study) and strongly negative europium anomalies (Eu* = 0.28). The REE trends of both Apollo 14 high-Al basalt 14053 and Apollo 15 KREEP basalt 15386 show LREE enrichment ($La/Sm_n$ = 1.96 and 2.30, respectively). However, in every other sense the trend for sample 14053 fits well with those of the Apollo 12 basalts, whereas KREEP basalt 15386 displays the highest REE abundances and the strongest negative europium anomaly (Eu* = 0.22)

among all the samples analysed in the present study. Generally, the high-Ti mare basalts display higher REE abundances than the low-Ti mare basalts.

In basaltic magmas, the large-ion lithophile elements (LILE - K, Rb, Cs), Sr, Ba, Zr, Hf, Nb, Ta, Th and U behave incompatibly in addition to the REEs. In our sample set, Apollo 15 KREEP basalt 15386 consistently displays the highest incompatible-element abundances, followed by the Apollo 11 high-K basalts (Table 3, Fig. 4). Apollo 14 high-Al basalt 14053 also contains elevated K, Rb, Cs, Ba, La, Th and U concentrations compared to the other mare basalts. Incompatible element bivariate plots not only highlight compositional variations between the high- and low-Ti mare basalts, but also show differences between the Apollo 11 high- and low-K basalts, and between the individual Apollo 17 basalt groups (Fig. 4). Apart from sample 15555,982, the Apollo 12 and 15 low-Ti basalts in our sample set show similar incompatible-element abundances. The uncharacteristically low incompatible-element abundances in sample 15555,982 support the conclusion that this sample has a heterogeneous composition (Ryder and Schuraytz, 2001; Spicuzza et al., 2007).

## 5. DISCUSSION OF PETROGRAPHY AND BULK-ROCK DATA

### 5.1. Apollo 12 and 15 Low-Ti basalts

Both our data, and that from previous studies (e.g., Kushiro and Haramura, 1971; Rhodes et al., 1977; Neal and Taylor, 1992; Neal et al., 1994a; Ryder and Schuraytz, 2001; Schnare et al., 2008), highlight similarities between the major element data of Apollo 12 and Apollo 15 low-Ti basalts (Fig. 1). Comparable REE and other incompatible element abundances in low-

444 Ti basalts from these two sites indicate that they originated from compositionally similar
445 parental melts. However, the slight compositional differences, which are present, as well as
446 differences in Nd isotopic data, indicate that these two basalt groups were not derived from
447 the same source (e.g., Neal and Taylor, 1992; Snyder et al, 2000). These differences also
448 apply to the three groups of Apollo 12 mare basalts – olivine, pigeonite and ilmenite – which
449 are also thought to have been produced by three separate source regions (e.g., Neal and
450 Taylor, 1992; Snyder et al., 1997; Snyder et al., 2000). The variations in bulk-rock REE
451 compositions and compatible trace element ratios among mare basalts of the same type (e.g.,
452 the Apollo 12 ilmenite basalts) highlight the role of crystal fractionation (and accumulation)
453 in the evolution of the parental melts. Increasing Sc abundance with decreasing Mg#
454 indicates olivine fractionation in Apollo 12 mare basalts, but this trend is not observed for
455 Apollo 15 mare basalts. In addition, ilmenite basalts 12051 and 12064 contain ≥ 11 wt %
456 $Al_2O_3$, a characteristic Dungan and Brown (1977) attributed to olivine fractionation, rather
457 than origin from a high-Al parental melt. The strong decrease in Cr with increasing Mg# in
458 Apollo 12 basalts is probably a result of Cr-rich spinel co-fractionation with olivine. Again
459 this trend is not observed in the Apollo 15 mare basalt data. Therefore, although the Apollo
460 12 and 15 mare basalts appear to have been produced from relatively similar parental melt
461 compositions, compatible trace element abundances suggest the melt evolution of these two
462 groups differed considerably.
463
464 Comparison of olivine forsterite content with bulk-rock Mg# reveals that olivine basalt 12040
465 contains a significant amount of accumulated olivine (appendix 1), in agreement with the data
466 of Meyer and Wilshire (1974), Baldridge et al. (1979), and Neal et al. (1994b). Olivine
467 accumulation can also explain this sample's high bulk-rock MgO and low $Al_2O_3$ and CaO
468 contents, and a lack of correlation between $Cr_2O_3$ and MgO (accumulated olivine grains are

not consistently associated with spinel grains). MgO enrichment, along with other major and trace element depletions, is shown, to a lesser extent, by ilmenite basalt 12016 (Table 3, Fig. 1). Neal et al. (1994a) suggested heterogeneous olivine distribution in this sample, and therefore, these characteristics indicate that 12016,37 originated from an olivine-rich area of the sample.

### 5.2. Apollo 11 and 17 High-Ti basalts

The modal mineralogy (Table 1) and bulk-rock major-element data from this and previous studies (Fig. 1), show that the Apollo 17 basalts extend to higher $TiO_2$ contents than the Apollo 11 basalts, with several types: B, C and unclassified samples containing over 11 wt% $TiO_2$ (e.g., Brunfelt et al., 1974; Hodges and Kushiro, 1974; Boynton et al., 1975). In contrast, the Apollo 11 mare basalts extend to higher $Al_2O_3$ contents than the Apollo 17 mare basalts (11 vs. 9.6 wt %, respectively). These consistent differences in major element compositions reflect compositional variations in the parental melts of different groups of high-Ti basalts (Table 1, Neal and Taylor, 1992 and references therein). The Apollo 17 basalts in our sample set represent five different basalt groups thought to originate from distinct parental melts (Table 1): types A, B and C, and two unclassified basalts, which do not appear to be related to each other or the other basalt types (Shih et al., 1975; Rhodes et al., 1976; Warner et al., 1979; Ryder, 1988, 1990). Apollo 11 type B3 basalt 10020 (see Table 1 for type characteristics) and Apollo 17 type A basalt 75055 appear to be the end-members in terms of major and trace element abundances, almost bridging the gap between Apollo 11 low-K and Apollo 17 basalt compositions. Neal et al. (1990a) also noted the relative similarity between these two basalt groups. Snyder et al. (1990) suggested that the Apollo 11

494 type B3 and Apollo 17 type C basalts share chemical characteristics, and, therefore, originate
495 from similar parental melts. Our bulk-rock REE abundances and Sm/Hf ratios within Apollo
496 11 type B3 basalt 10020 and Apollo 17 type C basalt 74275 are similar, but other trace
497 element and major element abundances do not suggest any close relationship between these
498 two samples. The Apollo 11 low-K basalts show varying trace element abundances, from
499 type B3 basalt 10020 with the highest Mg# (42) and lowest REE abundance (Sm = 9 ppm), to
500 type B1 basalt 10058 with the lowest Mg# (37) and highest REE abundance (Sm = 16 ppm)
501 (Table 3, Fig. 3). Despite this apparent co-magmatic evolution, these three samples are
502 members of separate basalt groups that cannot be related via direct shallow level crystal
503 fractionation (sample 10050 is an unclassified low-K basalt) - La/Sm ratios were found to
504 vary too widely at similar Co abundances (Beaty and Albee, 1978; Beaty et al., 1979a,b;
505 Rhodes and Blanchard, 1980). However, Snyder et al. (1992b, 1993) and Jerde et al. (1994)
506 suggest that groups B1 and B3 may have originated from the same source at different periods
507 in time.
508
509 Our mineralogical and chemical observations, along with previously published data (e.g.,
510 Kesson, 1975; Neal et al., 1990a; Paces et al., 1991; Neal and Taylor, 1992), suggest that the
511 high-Ti basalts in our sample set are affected by crystal fractionation/accumulation (as are the
512 low-Ti basalt samples). Decreasing V and Cr content with decreasing Mg# in the high-Ti
513 mare basalts suggests ilmenite and Cr-spinel fractionation in their parental magmas (V is
514 highly compatible in ilmenite). In general, the unclassified, type B(1 and 2) and C Apollo 17
515 basalts are enriched in V and Cr, compared to the Apollo 17 type A samples and Apollo 11
516 basalts (Table 3; Fig 2b,c). This is consistent with higher ilmenite and spinel modal
517 abundances in the former sample types (Table 2). As the Apollo 11 low-K and Apollo 17
518 basalts show similar REE and LILE contents, compatible element compositional differences

519 cannot be related to parental melt evolution in these samples (Apollo 11 high-K REE and
520 LILE content is discussed below). Therefore, the two basalt groups must have originated
521 from different parental melt compositions. The Apollo 17 type B2, C and the majority of the
522 unclassified basalts appear to have formed from a melt enriched in $TiO_2$, Cr and V, compared
523 to the Apollo 17 type A and Apollo 11 low-K basalts.
524
525 Major and trace element data collected in this study, along with that of previous studies (e.g.,
526 LSPET, 1969; Beaty and Albee, 1978; Neal and Taylor, 1992; Jerde et al., 1994), suggest that
527 all of the Apollo 11 high-K basalt samples originate from the same melt. The evolution of
528 this melt can be tracked in our samples – REE and other incompatible elements increase in
529 abundance from the most primitive sample (10072) to the most evolved sample (10049)
530 (Table 3, Fig. 4). Separation of the Apollo 11 low- and high-K basalts is not simply based on
531 differing potassium abundances. Incompatible elements, $P_2O_5$ and $Na_2O$ are more abundant
532 in the high-K basalts (Table 3, Figs. 3 and 4). Previous studies also highlight these
533 differences (e.g., Haskin et al., 1970; Philpotts and Schnetzler, 1970; Rhodes and Blanchard,
534 1980; Neal, 2001). The elevated abundances of K and P in the Apollo 11 high-K basalts,
535 along with strongly negative Eu anomalies (for all four samples analysed in this study Eu* =
536 0.28), and LREE enrichments compared with the low-K samples ($La/Sm_n$ = 0.66-0.85 and
537 1.27, respectively; Fig. 3), imply that these basalts have undergone KREEP contamination.
538 Neal and Taylor (1992) argued that if these basalts were the result of KREEP assimilation,
539 the final melt must have contained > 5 % assimilant to produce the observed K
540 concentrations. Assuming these samples originated from a melt similar to the Apollo 11 low-
541 K basalts, this amount of KREEP assimilation would have resulted in basalt samples with
542 strong LREE enrichments ($La/Sm_n$ ~2.30) characteristic of KREEP. As this enrichment is not
543 visible, either a KREEP component was present in the source (as suggested by Neal and

Taylor, 1992), or the parental melt composition was significantly different to that of the Apollo 11 low-K basalts (as suggested by Jerde et al., 1994).

### 5.3. Apollo 15 KREEP basalt and Apollo 14 low-Ti, high-Al basalt

In addition to containing higher amounts of incompatible trace elements, $K_2O$ and $P_2O_5$, Apollo 15 KREEP basalt 15386 is enriched in $Al_2O_3$, $SiO_2$ and $Na_2O$ compared with mare basalts (with the exception of some Apollo 17 VLT samples), and relatively poor in FeO (with the exception of some Apollo 14 VHK samples), MnO and Sc (Tables 1 and 3, Fig. 1-4). The Apollo 14 high-Al basalts, including sample 14053, are also $Al_2O_3$- and $K_2O$-rich, though both the Apollo 11 high-K samples and Apollo 15 KREEP basalts contain higher concentrations of these two elements (Fig. 1). Incompatible trace element abundances highlight the similarities between KREEP basalts and the Apollo 14 high-Al and Apollo 11 high-K mare basalts. Both Apollo 11 high-K basalts and Apollo 14 high-Al basalts show incompatible trace element abundances between those of the Apollo 15 KREEP basalts and the other mare basalts, with Apollo 11 high-K basalts always plotting closest to the KREEP compositions (Table 3, Fig. 4). Apollo 15 KREEP basalt 15386 shows a typical KREEPy REE signature (Warren, 1989), i.e., overall high abundance with strong LREE enrichment ($La/Sm_n = 2.30$), relative HREE depletion ($Gd/Lu_n = 1.44$), deeply negative Eu anomaly ($Eu^* = 0.22$), and a slightly convex upwards trend (Fig. 3). Previous studies have also reported highly enriched initial isotopic compositions for this sample (e.g., Shearer et al., 2006 and references therein). These distinctive characteristics are typical of a parental melt derived from a source region containing a component of residual melt from the very end of LMO solidification. Apollo 14 high-Al basalt 14053 shows LREE enrichment compared with the

569 HREE's ($La/Lu_n$ = 1.58), and does have a slightly convex upwards trend, but its REE pattern
570 is generally in line with the low-Ti mare basalts. The negative Eu anomaly of sample 14053
571 is also only slightly stronger than those of the low-Ti mare basalts analysed in this study (Eu*
572 = 0.49 and 0.56-0.66, respectively). From the above data and previous studies (see Fig. 1,
573 Apollo 14 references), it is clear that the Apollo 14 high-Al mare basalt melts were
574 contaminated with a material rich in incompatible elements, especially LREE and Rb.
575 However, because the Apollo 14 high-Al basalts do not appear to be as enriched in
576 incompatible trace elements as the Apollo 11 high-K basalts, either the contaminant was of a
577 different composition or the proportion of contamination was less in the former (Neal and
578 Kramer, 2006).
579
580 **5.4. The origin of mare basalts**
581
582 Compositional variation between the high- and low-Ti mare basalt parental melts is thought
583 to be related to different degrees of partial melting of distinct source regions (e.g., Ringwood
584 and Essene, 1970; Duncan et al., 1976; Nyquist et al., 1977, 1979; Snyder et al., 1992a;
585 Snyder et al., 1997). Previous studies indicate that the low-Ti basalts were produced from
586 olivine-pyroxene cumulate sources that formed during the first 78-80 % crystallisation of the
587 LMO, and that the high-Ti basalts formed as a result of such olivine-pyroxene cumulates
588 mixing with late stage (≥ 87 % LMO crystallisation) ilmenite-rich cumulates (Ringwood and
589 Kesson, 1976; Ryder, 1991; Spera, 1992; Snyder et al., 1992a; Elkins-Tanton et al., 2002;
590 2011). As the ilmenite-rich cumulates formed during a late stage of LMO solidification, it
591 seems probable that some instantaneous residual liquid would have been trapped during
592 ilmenite-rich cumulate crystallisation, which could explain the relatively high REE
593 abundances in high-Ti mare basalts. However, previous studies indicate that this trapped

instantaneous residual liquid (TIRL) was present in both ilmenite-rich and olivine-pyroxene cumulate source regions (Snyder et al., 1992a, 1994; 1997), although TIRL in earlier cumulates would have been less REE enriched. In addition, subsequent LMO overturn is believed to have mixed the two cumulate source regions to some degree, but it is unlikely that this process completely homogenised the LMO products (e.g., Ringwood and Kesson, 1976; Binder, 1985; Huges et al., 1990; Snyder et al., 1992a; Elkins-Tanton et al., 2002; 2011). The amount of trapped KREEPy liquid in a source, along with different degrees of partial melting, would significantly affect the REE composition of a mare basalt melt. This parental melt would subsequently evolve via crystal fractionation/accumulation and possible assimilation prior to final crystallisation. Therefore, chemical modelling is required to trace the REE patterns in a given mare basalt sample back to a possible source region composition. Via this modelling, the mineralogical and chemical composition of the source region of each mare basalt type within our sample set can be estimated and compared, allowing an investigation of the level of heterogeneity in the lunar interior.

## 6. CHEMICAL MODELLING

The main geological processes controlling the chemical composition of igneous rocks are partial melting of the source region (mantle), magma ascent, fractional crystallisation, crystal accumulation, magma mixing and contamination, all of which affect bulk-rock trace element abundances. Therefore, by comparing the bulk REE composition of a sample with theoretically calculated values, an informed estimate of its petrological history can be ascertained. We used forward REE geochemical modelling to determine if, and how, each

mare basalt type could have evolved from partial melting of specific low- and high-Ti cumulate sources.

### 6.1. Mare basalt parental melt compositions

Melt evolution commonly alters the composition of a basaltic melt prior to its complete crystallisation. Therefore, the REE pattern of even the most primitive sample may not be representative of the parental melt. To overcome some of these issues, the composition of a number of samples may be averaged to minimise the effects of crystal accumulation/fractionation, and reconstruct primary elemental abundances (e.g., Neal et al., 1994b; Snyder et al., 1992a, 1997). Alternatively, petrogenetic modelling can be used to calculate the probable parental melt composition (e.g., Neal et al., 1990a). Table 4 shows the estimated parental melt REE compositions for the mare basalt groups represented in our sample set, and the method by which they were calculated.

If the parental melt is taken as a starting composition, it can be used to indicate the extent of crystal accumulation and/or fractionation involved in the petrogenesis of rock samples within the same group. Samples 12040, 12016, 15016 and 74275 all show lower REE abundances than their inferred parental melts (Fig. 5), which, in conjunction with evidence from compatible element abundances and mineral modes (appendix 1), indicates olivine accumulation in these samples. Apollo 12 ilmenite basalt 12064 and Apollo 17 type A basalt 75055 also show lower REE abundances than their parental melts, but as they contain little to no olivine, and ilmenite modal abundances are not atypically high (Table 1; Brown et al.,

1975; Neal et al., 1994a), these REE patterns probably indicate small amounts of clinopyroxene accumulation.

The remaining mare basalts in our sample set (excluding unclassified samples 10050, 70017 and 70035, as their parental melt compositions cannot be estimated) contain REE abundances similar to their parental melts, or have undergone some crystal fractionation. As sample 10058 is the most primitive of only three basalts in the Apollo 11 low-K type B1 basalt group (e.g., Beaty and Albee, 1978; Rhodes et al., 1980), its parental melt composition cannot be accurately estimated without using experimental petrology. Therefore, this sample has not been included in our modelling.

### 6.2. Selecting an appropriate mare basalt source composition

Assuming the parental melt REE compositions listed in Table 4 represent primary melts (i.e. nothing has happened to significantly change the REE composition between source partial melting and parental melt production), they can be used to calculate the amount of partial melting that is required to produce each basalt group, if appropriate REE and mineral compositions are assumed for the parental-melt source region(s). The chemical modelling of Snyder et al. (1992a) assumed an initial LMO source composition of 3× chondritic values for the REE, following the rational of Hughes et al. (1988, 1989). This modelling indicates that after 78 % LMO crystallisation a negative Eu anomaly is produced in the residual liquid, because plagioclase becomes a liquidus phase at this point. As mare basalt bulk-rock REE patterns show a negative Eu anomaly, their source regions must have formed after plagioclase appeared on the liquidus during LMO crystallisation. Therefore, for this study, the Snyder et

667 al. (1992a) cumulate composition at 86 % crystallised solid (PCS) with the addition of 1-2 %
668 trapped instantaneous residual liquid (TIRL) was chosen as an appropriate source region
669 composition for the low-Ti basalts (Table 5). TIRL was included because, based on Lu/Hf
670 and Sm/Nd isotope ratios, Beard et al. (1998) calculated that most of the low-Ti and the high-
671 Ti mare basalt source regions contained a small proportion of residual trapped liquid, in
672 agreement with the models of Snyder et al. (1992a; 1994, 1997). As a small proportion of a
673 high-REE component is also required in the production of the low-Ti picritic glass beads
674 (e.g., Elkins-Tanton et al., 2003), a proportion of residual liquid was likely involved in partial
675 melts of the LMO mafic cumulates. The REE composition of the high-Ti source region was
676 assumed to be an 80:20 mixture of the 86 PCS cumulate composition with more evolved 95
677 PCS cumulate composition, as well as the addition of 1 % TIRL from the 95 PCS level
678 (Table 5) (Snyder et al. 1994).
679
680 In terms of mineralogy, recent numerical modelling of LMO crystallisation suggests that 50
681 % equilibrium crystallisation produces a lower mantle consisting of mostly olivine, with
682 minor orthopyroxene (7-8 %), and possible Cr-spinel and/or garnet (~3 %) depending on
683 depth (Elardo et al., 2011). As crystallisation continues orthopyroxene becomes the dominant
684 crystallising phase, until plagioclase and clinopyroxene begin to crystallise (at ~ 78 % and 86
685 % LMO crystallisation, respectively) (Snyder et al., 1992a). Most plagioclase crystals would
686 have been removed via flotation to form the anorthositic primary crust. Snyder et al. (1992a)
687 calculated that pigeonite rather than orthopyroxene would crystallise after 78 PCS. Therefore,
688 low-Ti mare basalt source regions with an 86 PCS cumulate composition should consist of
689 mostly olivine, orthopyroxene and pigeonite, possibly with minor (<5 %) clinopyroxene,
690 spinel/garnet and some retained plagioclase. This mineralogy can be used as a good
691 approximation for the low-Ti mare basalt source. Utilising geochemical modelling, Snyder et

al. (1992a) calculated that the high-Ti mare basalt source would have an Mg# of 0.73, and a modal mineralogy of 46 % pigeonite, 43 % olivine, 7 % clinopyroxene, 3 % ilmenite and 1 % plagioclase. Based on the more recent data of Elardo et al. (2011), we have modified this mineralogy to include orthopyroxene as a replacement for half of the pigeonite (to give 23 % of each). However, this modification makes minimal difference to melt-REE evolution, as the partition coefficients of orthopyroxene and pigeonite are very similar (Table 6).

### 6.3. Fractional and batch partial melting

The two end-members of the partial melting processes are batch melting and fractional melting, but in reality partial melting is generally a combination of the two. The dominant process can be revealed by calculating the progression of REE compositions in a melt for 100 % batch and 100 % fractional melting, and comparing these compositions to those observed in the parental melt. Where modal melting occurs (i.e., each mineral phase melts in proportion to its modal abundance in the source), batch melting can be calculated using the following equation:

$$C_L/C_0 = 1/[D_0 + F(1-D_0)] \quad (1)$$

where; $C_L$ = weight concentration of a trace element in the liquid

$C_0$ = weight concentration of a trace element in the original unmelted source

F = weight fraction of melt produced

$D_0$ = bulk distribution coefficient of the original solid.

717 The bulk distribution coefficient can be calculated by multiplying each mineral partition
718 coefficient by the fraction of that mineral in the source. We used the REE partition
719 coefficients of McKay (1986) for olivine, Yao et al. (2012) for orthopyroxene and augite,
720 McKay et al. (1991) for pigeonite, Phinney and Morrison (1990) for plagioclase, Nakamura
721 et al. (1986), Klemme et al. (2006) and van Kan Parker et al. (2011) for ilmenite, and Green
722 et al. (2000) for garnet (Table 6). The values of McKay (1986), McKay et al. (1991), Phinney
723 and Morrison (1990) and Nakamura et al. (1986) were also used by Snyder et al. (1992a;
724 1995) as the most relevant for lunar magmatic conditions, and were more recently used by
725 Neal and Kramer (2006). The plagioclase partition coefficients of Hui et al. (2011) are mostly
726 in agreement with those of Phinney and Morrison (1990). The latter dataset was selected for
727 this study above the former as it is more complete for the REE. The recent datasets of Yao et
728 al. (2012), Klemme et al. (2006) and van Kan Parker et al. (2011), were all produced with
729 parameters appropriate to a lunar environment (anhydrous conditions, oxygen fugacity at IW
730 +1.5, basaltic/peridotitic melts). The more classic datasets mentioned above were produced
731 under similar conditions (where specified).
732
733 Fractional melting can be represented by equation 2, again assuming each mineral phase
734 melts in proportion to its modal abundance in the source:
735
736 $$C_L/C_0 = (1/D_0) \times (1 - F)^{(1/D_0-1)} \quad (2)$$
737
738 Using the cumulate source compositions of Snyder et al. (1992a) and Snyder et al. (1994)
739 (Table 5) as approximations of the low- and high-Ti mare basalt sources ($C_0$), the low- and
740 high-Ti mare basalt source region modal mineralogies calculated by Elardo et al. (2011) and
741 Snyder et al. (1992a), and the mineral partition coefficients mentioned above ($D_0$) (Table 6),

742 the weight concentration of REE in the liquid ($C_L$) can be calculated for increasing melt
743 fractions (F).
744
745 Snyder et al. (1997) argued that mare basalt partial melting was non-modal, as the trapped
746 instantaneous residual liquid (TIRL) would have been the first phase to melt. These authors
747 stated that only once this liquid had completely melted would other phases begin to melt. As
748 this liquid cannot be ascribed to a single mineral phase, it cannot be included in Eqns. 1 and
749 2. However, as previously stated, to simulate its effect on partial-melt REE composition, a
750 small percentage (1 %) of the TIRL in equilibrium at 86 PCS and 95 PCS was included in the
751 low- and high-Ti mare basalt source region compositions, respectively (Table 5).
752
753
754 *6.3.1. Low-Ti mare basalts.*
755
756 Our partial melting calculations suggest that 1-3 % fractional melting of the assumed low-Ti
757 source would yield similar REE abundances to those observed in the parental melts of Apollo
758 12 pigeonite and olivine basalts (Fig. 6a). The best fit is produced by assuming the following
759 modal mineralogy for the source: 52 % olivine, 23 % orthopyroxene, 23 % pigeonite and 2 %
760 clinopyroxene. No plagioclase is required in the source, although the addition of up to 1 %
761 does not greatly affect the REE source composition (Snyder et al. 1992a), or the evolution of
762 the melt. The Apollo 12 ilmenite basalts contain slightly higher REE abundances, matching
763 with melts derived from 1-2 % fractional melting of the low-Ti source (Fig 6b). However, in
764 this case 1 % plagioclase is required in the source to reproduce the Eu anomalies. In addition,
765 the best fit is produced if clinopyroxene is absent from the source, as even a 1 % level
766 abundance of this phase decreases the calculated $La/Lu_n$ ratio beyond that of the measured

767 data. In contrast, the chemical modelling results of Snyder et al. (1997) suggest the Apollo 12
768 mare basalt sources do contain clinopyroxene, 30 % in the olivine-pigeonite basalt source and
769 11.5 % in the ilmenite basalt source. Our partial melting estimates are also lower than those
770 of Snyder et al. (1997), who calculated that the Apollo 12 olivine and pigeonite basalts could
771 be produced by 7-9 % partial melting, and the ilmenite basalts could be produced by 5-7 %
772 partial melting. The discrepancy is probably produced by a combination of differing chemical
773 modelling techniques and assumed source TIRL percentages. As mentioned above, the
774 chemical modelling of this study attempts to reproduce mare basalt partial melt REE
775 compositions by assuming 1 % TIRL in the mare basalt source prior to modal melting.
776 Snyder et al. (1997) aimed to recreate the effects of non-modal melting by adding TIRL after
777 the partial melting compositions were produced, assuming the TIRL would always melt first.
778 For example, assuming 1 % TIRL, a 2 % partial melt would contain one-half TIRL, a 5 %
779 partial melt would contain one-fifth TIRL, etc. This modelling method gave Snyder et al.
780 (1997) greater freedom to experiment with the percentage of TIRL added to each source,
781 rather than relying on limited previously published source compositions. The Sm-Nd and Rb-
782 Sr data of Snyder et al. (1997) and Snyder et al. (2000) indicate that the Apollo 12 olivine-
783 pigeonite basalt source contained <1 % TIRL, and that the ilmenite basalt source contained
784 ~0.5-0.2 % TIRL. These percentages are adopted in the chemical modelling of Snyder et al.
785 (1997). A lower percentage of TIRL in the source would decrease the REE abundance in a
786 given modal partial melt, especially for the LREE (Fig 6a,b). However, the non-modal
787 melting calculations of Snyder et al. (1997) appear to more than compensate for this affect.
788 Based on Sm-Nd and Rb-Sr data, the assumed source of the present study over represents
789 TIRL for the Apollo 12 basalts. However, our data do show that a 1 % TIRL source produces
790 a much closer match to the measured parental melts than a 0 % TIRL source, thus

791 demonstrating the strong affect a small amount of TIRL can have on source region REE
792 abundances.
793
794 In contrast to the Apollo 12 basalts, the Apollo 15 olivine- and quartz normative mare basalts
795 appear to originate from a source with ~2 % TIRL (Snyder et al., 2000). The REE
796 abundances of a source with 2 % TIRL were calculated by Snyder et al. (1992a), therefore we
797 can compare partial melts from sources with both 1 and 2 % TIRL for the Apollo 15 olivine
798 normative basalts. Assuming 1 % TIRL in the low-Ti source, REE compositions of the
799 Apollo 15 olivine-normative parental melt roughly correspond to 1-4 % batch melting, if the
800 source mineralogy is assumed to be the same as for Apollo 12 olivine and pigeonite basalts
801 (Fig. 6c). 3-9 % batch melting of a source with 2 % TIRL also provides an approximation of
802 the measured Apollo 15 olivine-normative parental melt value. However, the chondrite
803 normalised La/Lu ratio of the measured parental melt (2.42) is higher than the calculated
804 values (1.47-0.54 and 2.19-1.17 for the 1 % and 2 % TIRL sources, respectively). The
805 difference between measured and calculated $La/Lu_n$ ratios is reduced if 0.5 % garnet is added
806 to the source region in our models (Fig. 6d). The presence of garnet in the low-Ti mare basalt
807 source regions was previously suggested by Khan et al. (2006) as a way of explaining high
808 Sm/Yb ratios. In addition, Beard et al. (1998) proposed that it could explain the Lu-Hf ratios,
809 and Hf isotope systematics. However, as our low-Ti mare basalt source region REE
810 compositions are set to those of Snyder et al. (1992a), the effect of garnet on REE
811 composition of the source could not be included in our models. Therefore, any conclusions
812 regarding garnet in the source of Apollo 15 olivine-normative basalts remain equivocal.
813

814 *6.3.2. High-Ti mare basalts.*

815

816 The Apollo 11 low-K mare basalt (type B3) parental melt contains REE abundances similar
817 to a 3-5 % fractional melt of the assumed high-Ti source (Fig. 6) – an 80:20 mixture of 86
818 and 95 PCS cumulates with 1 % TIRL (Snyder et al., 1994). This is in agreement with Snyder
819 et al. (1992a), who suggested that the Apollo 11 low-K basalts were produced by 1-3 %
820 partial melting, also in agreement with Ringwood and Essene (1970). The measured and
821 calculated REE trends match well if clinopyroxene is decreased to 2 % in the source region
822 and olivine is correspondingly increased, compared to the suggested mineral modes of the
823 source region by Snyder et al. (1992a) (7% clinopyroxene and 43% olivine). As with the
824 Apollo 12 modelling comparisons, this necessary reduction in clinopyroxene content may be
825 explained by differing modelling techniques and possibly <1 % TIRL in the source (although
826 Snyder et al. (2000) indicate the Apollo 11 B3 basalts contain close to 1 % TIRL).

827

828 The Apollo 11 B3 basalt source mineralogy can also be successfully applied for the
829 production of the parental melts of Apollo 17 basalts, although melting proportions and
830 dominant melting end-members vary. Apollo 17 type A basalts show parental melt REE
831 composition corresponding to 4-6 % fractional melting of the assumed high-Ti mare basalt
832 source, whereas type B2 basalts correspond to 3-6 % batch melting, and type C basalts
833 correspond to 2-4 % fractional melting (Table 7; Fig.6). These calculations are in agreement
834 with Duncan et al. (1976), who demonstrated that the Apollo 17 high-Ti basalts were
835 produced by < 20 % partial melting. However, even with reduced clinopyroxene content in
836 the source region, for Apollo 17 type A basalt, the measured Eu anomaly is smaller than the
837 calculated value for the source (Eu* = 0.60 in sample 75055,120, and 0.15 in the calculated
838 source region). This may be the result of less than 1 % TIRL in the type A source region. A

decreased amount of TIRL in the source would produce a weaker Eu anomaly, and decrease the amount of partial melting required to produce the parental melt. Alternatively, the type A basalts at the Apollo 17 site may have been produced from a source with a different cumulate component, for example, the source may have been a mixture of 86 PCS and 90 PCS cumulates, rather than 95 PCS. A source of this type would have a weaker Eu anomaly (Snyder et al., 1992a).

## 6.4. Hybrid source compositions

### *6.4.1. Apollo high-K basalts*

As previously stated, the Apollo 11 high-K basalts (type A) contain high levels of REEs, and strongly negative Eu anomalies, compared to the other high-Ti samples. Jerde et al. (1994) argued that the high-K basalt compositions are consistent with formation from a parental liquid with the composition of Apollo 11 orange glass (in agreement with the fractional crystallisation models of Longhi (1987)), coupled with 7.5-15 % assimilation of a KREEPy material. These calculations are also in agreement with Hughes et al. (1989), who suggested a 10 % KREEPy contaminant for the Apollo 11 high-K basalts. We attempted to model the REE compositions produced by high-K KREEP (Warren, 1989) assimilation in this parental melt using assimilation and fractional crystallisation (AFC) calculations (Eqn. 4).

$$C_L/C_0 = f' + (r/(r - 1 + D)) \times ((C_A/C_0) \times (1 - f')) \quad (4)$$

where; $C_L$, $C_0$, D and F represent the same factors as in Eqn. 3,

$r$ is the ratio of the assimilation rate to the fractional crystallisation rate,

865 $C_A$ is the concentration of the trace element in the assimilated wall rock,

866 and *f'* is represented by Eqn. 5.

867

868 $$f' = F^{-(r - 1 + D)/(r - 1)} \quad (5)$$

869

870 The results of these calculations suggest that after 10 % assimilation/crystal fractionation of
871 high-K KREEP and a 50 % ilmenite, 30 % pigeonite and 20 % augite fractionating
872 assemblage, with a ratio (*r*) of 0.8, a parental melt with the composition of Apollo 11 orange
873 glass (Table 4) could have produced similar REE abundances to those observed in the Apollo
874 11 high-K basalts. However, most authors now agree that there is no direct parent-daughter
875 relationship between the picritic glasses and mare basalt melts (e.g., Longhi 1992a; Papike et
876 al., 1998), and that the glasses originate from deeper source regions (see Neal et al. 2001, and
877 Shearer et al., 2006, and references in both). In addition, anything greater than 5 % high-K
878 KREEP assimilation would increase the $La/Sm_n$ ratio above the ratios measured in the Apollo
879 11 high-K basalts (e.g., Neal and Taylor, 1992), as demonstrated in Fig. 7a. Therefore, not
880 only is it probable that the Apollo 11 high-K basalts, like the other high-Ti mare basalts,
881 originated from a cumulate source similar to that modelled by Snyder et al. (1994) (Table 5),
882 but also that KREEP plays a limited role in the origin of these basalts. To test this theory, we
883 attempted to model the evolution of Apollo high-K basalt REE's from a cumulate source with
884 < 5% high-K KREEP assimilation.

885

886 Various elemental and isotopic ratios indicate that the Apollo 11 low- and high-K basalts did
887 not originate from the same source (e.g., Neal and Taylor, 1992; Shearer et al. 2006).
888 Notably, the two types display different εNd values (deviation of $^{143}Nd/^{144}Nd$ from
889 chondritic), indicating different amounts of TIRL in each source (Snyder et al. 2000). In

accordance with these values, 2 % TIRL from the 95 PCS level was added to the assumed high-Ti cumulate source (80:20 mixture of 86 and 95 PCS cumulate REE compositions). For these calculations, re-calculated TIRL data, based on the models of Snyder et al. (1992a), were used (J. Davenport, pers. comm.). Our calculations indicate that a 5 % fractional melt of this source, with the same mineralogy as that of the modelled Apollo low-K source, could produce REE composition similar to those of the Apollo 11 high-K basalts with only 4 % total high-K KREEP assimilation. Despite this, the $La/Sm_n$ ratios of modelled vs. measured values still do not match satisfactorily (1.03 and 1.27-1.28, respectively). This may indicate that the Apollo 11 high-K assimilant had a lower $La/Sm_n$ ratio, which would be the case for a less evolved material than high-K KREEP (< 99 PCS). However, this material would have to be evolved enough to produce the strongly negative Eu anomalies measured. This scenario is supported by the data of Gaffney et al. (2007), who reported depleted initial Nd and Sr isotopic compositions in high-K basalt 10017; suggesting KREEP was not involved in the formation of this sample.

*6.4.2. Apollo 14 high-Al basalts*

Most authors agree that the production of the Apollo 14 high-Al basalts included a certain proportion of assimilation, and Neal and Kramer (2006) suggested the inclusion of a granitic component. These authors state that the Apollo 14 high-Al basalts of groups B and C (the latter containing sample 14053) acquired elevated LREE concentrations through < 5 % assimilation of a mixture of KREEP and granitic material. However, neither KREEP nor granite is Al-rich, implying the high Al signature in these basalts was inherited from the source. Hughes et al. (1990) suggested that the Apollo 14 high-Al mare basalt source-region contained up to 3 % plagioclase - a potential source of Al. This hypothesis is supported by the

misfit produced in our AFC calculation when a low-Al, low-Ti mare basalt source is assumed (86 PCS and 1 % TIRL from the 95 PCS level, Snyder et al. 1992a). For these calculations to produce REE values remotely similar to those found in sample 14053, the granitic component of Neal and Kramer (2006) must be assimilated into a 1 % batch melt of the source at low *r* (0.1). Under these conditions 40 % crystal fractionation of a purely olivine assemblage is required, resulting in 2 % total assimilation (Fig. 7c). However, the measured vs. calculated $La/Lu_n$ ratios in this model do not match well. This issue can be corrected by the addition of plagioclase to the source. If the source region composition (Table 5) and mineralogy of Hughes et al. (1990) is assumed (92.9 % olivine, 2 % clinopyroxene. 3.1 % plagioclase, 0.77 % pigeonite, 0.51 % ilmenite, and 0.76 % TIRL at the 98 % LMO crystallisation level), the REE abundances of sample 14053 can be recreated more accurately. AFC calculations indicate that 40 % crystal fractionation of olivine from a 5 % batch melt of this source could produce similar REE abundances to those in sample 14053 (Fig. 7d), assuming the addition of a purely granitic assimilant (Table 5) and low *r* (0.21). Addition of a KREEP component to the assimilant, as suggested by Neal and Kramer (2006), results in a strongly negative Eu anomaly and a higher $La/Lu_n$ ratio than that measured in sample 14053. To counteract the effects of the more KREEP-rich Neal and Kramer (2006) assimilant the value of *r* must be decreased to 0.05, and the amount of crystal fractionation increased to 60 %. Therefore, if a relatively plagioclase-rich source is assumed, a purely granitic assimilant was probably involved in the formation of sample 14053 and the other group C (and possibly B) high-Al Apollo 14 basalts.

## 7. CHEMICAL MODELLING CONCLUSIONS

Modelling the evolution of REE from mare basalt source to sample involves many assumptions, not least of which are the composition of the source region and primary melts produced from the source. Previous attempts to model mare basalt source region composition, such as those of Snyder et al. (1992a, 1994, 1997), by necessity involved making assumptions about bulk Moon composition. Therefore, our models are by no means definitive, but we can draw some broad and important conclusions based on the results presented in this study:

- The mare basalts were the product of small-degrees of partial melting (< 10 %) of the LMO cumulates.
- Neither batch nor fractional melting appears to dominate the sample set as a whole, although certain parental melts do exhibit the characteristics of one or the other. The Apollo 12, Apollo 11, plus Apollo 17 type A, and C parental melts appear to be dominated by fractional melting, whereas the Apollo 15 olivine-normative and Apollo 17 type B2 parental melts are dominated by batch melting.
- No single source mineralogy can be applied to all mare basalts, even within the low- and high-Ti subsets. Taking into account the fact that the mare basalts are known to originate from similar mantle depths (e.g., Rhodes et al., 1977; Neal and Taylor, 1992; Beard et al., 1998; Neal and Kramer, 2006), this variation is indicative of a laterally heterogeneous lunar mantle.
- Assuming parental melts are a good approximation of the primary melt REE composition for each basalt group, plagioclase is not required in the source of most mare basalts, with the notable exception of the Apollo 14 high-Al basalts. Addition of more than 1 % plagioclase to the source of other basalts produces weaker negative Eu

anomalies than those observed in the samples. However, the possibility remains that at some point between primary melt and parental melt production the negative Eu anomaly in any or all of these basalt groups was enhanced by plagioclase fractionation and/or KREEP assimilation.

- AFC calculations emphasize the compositional differences for possible assimilants between the Apollo 14 high-Al and Apollo 11 high-K mare basalts, highlighting the heterogeneity of the lunar crust.

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1561

**Table 1:** Samples analysed in the present study, along with a summary of their main petrological characteristics and the publications that aided in their classification.

| Samples | Mission | Basalt Type | Basalt Type Characteristics | References |
|---|---|---|---|---|
| 10017<br>10049<br>10057<br>10072 | Apollo 11 | High-Ti, Low-Al, High-K: Type A | > 2000 ppm $K_2O$ plus elevated incompatible element (particularly La, 20-40 ppm) and $P_2O_5$ contents. ~3.6 Ga. | Geiss et al. (1977)<br>Papanastassiou et al. (1977)<br>Haskin et al. (1970)<br>Philpotts and Schnetzler (1970)<br>Beaty and Albee (1978)<br>Beaty et al. (1979a,b)<br>Ma et al. (1980)<br>Rhodes and Blanchard (1980)<br>Neal and Taylor (1992)<br>Neal (2001) |
| 10058 | Apollo 11 | High-Ti, Low-Al, Low-K: Type B1 | Intermediate K (< 1000 ppm), low La (<10 ppm), ~3.72 Ga. | Geiss et al. (1977)<br>Papanastassiou et al. (1977)<br>Beaty and Albee (1978)<br>Beaty et al. (1978a,b)<br>Guggisberg et al. (1979)<br>Ma et al. (1980)<br>Rhodes and Blanchard (1980)<br>Neal and Taylor (1992) |
| 10050 | Apollo 11 | High-Ti, Low-Al, Low-K: Type U | Unclassified, does not fit with the other Apollo 11 low-K samples. | Geiss et al. (1977)<br>Beaty and Albee (1978)<br>Guggisberg et al. (1979)<br>Rhodes and Blanchard (1980) |
| 10020 | Apollo 11 | High-Ti, Low-Al, Low-K: Type B3 | low K (< 750 ppm), low La (<10 ppm), ~3.76 Ga. | Geiss et al. (1977)<br>Papanastassiou et al. (1977)<br>Beaty and Albee (1978)<br>Beaty et al. (1978a,b)<br>Guggisberg et al. (1979)<br>Ma et al. (1980)<br>Rhodes and Blanchard (1980)<br>Neal and Taylor (1992) |
| 12040 | Apollo 12 | Low-Ti, Low-Al, Low-K:<br>Olivine basalt | Accumulated olivine results in Mg# >45, Rb/Sr ratio is >0.008, $TiO_2$ is low (~2.0 - 3.5 wt %). | Compston et al. (1971)<br>Papanastassiou and Weasserburg (1971a)<br>James and Wright (1972)<br>Walker et al. (1976)<br>Rhodes et al. (1977)<br>Baldridge et al. (1979) |
| 12052 | Apollo 12 | Low-Ti, Low-Al, Low-K:<br>Pigeonite basalt | Mg# <45, Rb/Sr ratio is >0.008, $TiO_2$ is intermediate (~3.0 - 5.0 wt %). | Compston et al. (1971)<br>Papanastassiou and Weasserburg (1971a)<br>James and Wright (1972)<br>Walker et al. (1976)<br>Rhodes et al. (1977)<br>Baldridge et al. (1979) |
| 12016<br>12047<br>12051<br>12064 | Apollo 12 | Low-Ti, Low-Al, Low-K:<br>Ilmenite basalt | Mg# is variable, Rb/Sr ratio is <0.008, $TiO_2$ is high (~3.0 - 6.0 wt %). | Compston et al. (1971)<br>Papanastassiou and Weasserburg (1971a)<br>James and Wright (1972)<br>Walker et al. (1976)<br>Rhodes et al. (1977)<br>Baldridge et al. (1979) |
| 14053 | Apollo 14 | Low-Ti, High-Al, Low-K: Group C (groups A and B are not represented within our sample set). | 14 -11 wt % $Al_2O_3$, <0.3 wt % $K_2O$, K/La ratio ~ 100, age ~ 4.3 Ga. | Warner et al. (1980)<br>Dickinson et al. (1985)<br>Shervais et al. (1985a)<br>Neal et al. (1988a, 1989) |
| 15016<br>15555 | Apollo 15 | Low-Ti, Low-Al, Low-K:<br>Olivine Normative | 18 - 24 wt % FeO, 8 - 17 wt % MgO, La < 6.5 ppm, Sm < 4.5 ppm, ~3.2 Ga. | Chappell et al. (1972)<br>Maxwell et al. (1972)<br>Rhodes and Hubbard (1973)<br>Chappell and Green (1973)<br>Cuttitta et al. (1973)<br>Helmke et al. (1973)<br>Nava (1974)<br>Nyquist (1977) |
| 15386 | Apollo 15 | KREEP | 18 - 24 wt % FeO, 8 - 17 wt % MgO, La < 6.5 ppm, Sm < 4.5 ppm, ~3.2 Ga. | Chappell et al. (1972)<br>Maxwell et al. (1972)<br>Rhodes and Hubbard (1973)<br>Chappell and Green (1973)<br>Cuttitta et al. (1973)<br>Helmke et al. (1973)<br>Nava (1974)<br>Nyquist (1977) |
| 75055 | Apollo 17 | High-Ti, Low-Al, Low-K: Type A | Similar major element abundances to Type B, but 50 - 60 % higher incompatible trace element abundances | Rhodes et al. (1976)<br>Warner et al. (1979)<br>Neal et al. (1990a,b) |
| 70215 | Apollo 17 | High-Ti, Low-Al, Low-K: Type B(2) (type B1 is not represented within our sample set) | Similar major element abundances to Type A, but 50 - 60 % lower incompatible trace element abundances. Can be divided into type B1 and B2, where B2 contains slightly higher La ppm | Rhodes et al. (1976)<br>Warner et al. (1979)<br>Neal et al. (1990a,b) |
| 74275 | Apollo 17 | High-Ti, Low-Al, Low-K: Type C | Higher MgO and $Cr_2O_3$ contents than Type A and B, REE abundances similar to Type A, contain high Fo olivines ($Fo_{80}$). | Rhodes et al. (1976)<br>Warner et al. (1979)<br>Neal et al. (1990a,b) |
| 70017<br>70035 | Apollo 17 | High-Ti, Low-Al, Low-K: Type U | Unclassified, these samples do not fit into any of the above Apollo 17 groups, but neither do they group together. | Longhi et al. (1974)<br>Nyquist et al. (1974)<br>Brown et al. (1975)<br>Dickinson et al. (1989) |

**Table 2:** Modal mineralogy for basalts analysed in this study.

| Sample[a] | Pyroxene[b] | Plagioclase[c] | Olivine[d] | Ilmenite[e] | Spinel[f] | Armalcolite[g] | Silica[h] | Troilite[i] | Mesostasis[j] | Phosphates[k] | Sum |
|---|---|---|---|---|---|---|---|---|---|---|---|
| A11: 10017,372 | 47 | 29 | | 10 | | | | 0.06 | 14.19 | trace | 100 |
| 10020,232 | 48 | 33 | 5 | 14 | 0.13* | | | trace | trace | trace | 100 |
| 10049,94 | 50 | 33 | trace | 15 | | | 1.40 | 0.40 | trace | trace | 100 |
| 10050,168 | 51 | 33 | 1 | 14 | | | 1.55 | 0.17 | 0.10 | trace | 100 |
| 10057,64 | 56 | 36 | trace | 8 | | | | 0.04 | trace | trace | 100 |
| 10058,254 | 51 | 32 | | 11 | | | 4.92 | 0.35 | trace | trace | 100 |
| 10072,40 | 45 | 39 | 1 | 15 | | | | 0.03 | trace | trace | 100 |
| A12: 12016,37 | 51 | 31 | 14 | 4 | 0.57 | | | 0.02 | trace | | 100 |
| 12040,206 | 55 | 19 | 22 | 3 | 0.14 | | | 0.05 | trace | trace | 100 |
| 12047,34 | 51 | 39 | 1 | 6 | 0.25 | | 2.95 | 0.35 | | trace | 100 |
| 12051,229 | 60 | 30 | | 7 | 0.07 | | 2.84 | 0.26 | | trace | 100 |
| 12052,39 | 62 | 32 | 2 | 3 | 0.29 | | 1.37 | 0.22 | | | 100 |
| 12064,138 | 62 | 27 | | 5 | 0.54 | | 3.82 | 0.11 | 1.21 | 0.27 | 100 |
| A14: 14053,260 | 59 | 34 | 3 | 2 | 0.26 | | 0.92 | 0.13 | trace | trace | 100 |
| A15: 15016,223 | 61 | 23 | 12 | 1 | 2.83 | | | trace | trace | trace | 100 |
| 15386,54 | 33 | 51 | | 3 | 0.01 | | 2.68 | 0.11 | 10.50 | 0.58 | 100 |
| 15555,982 | 56 | 31 | 11 | 1 | 0.64 | | 0.52 | trace | trace | trace | 100 |
| A17: 70017,543 | 53 | 28 | 1 | 17 | 0.06 | 0.20 | 0.32 | 0.09 | trace | 0.03 | 100 |
| 70035,194 | 52 | 29 | | 18 | trace | 0.21 | 1.39 | 0.12 | trace | trace | 100 |
| 70215,326 | 63 | 12 | 7 | 17 | 0.08 | 0.11 | 0.02 | 0.02 | | | 100 |
| 74275,323 | 51 | 22 | 9 | 15 | 1.86 | 0.93 | 0.28 | 0.41 | | | 100 |
| 75055,120 | 55 | 31 | | 10 | | | 3.62 | 0.11 | trace | trace | 100 |

[a]Numbers beginning with 10, 12, 14, 15 and 70 represent samples from the Apollo 11, 12, 14, 15 and 17 sites respectively. [b]pyroxene = clinopyroxenes of the solid solution system diopside ($CaMgSi_2O_6$) - hedenbergite ($CaFeSi_2O_6$) - enstatite ($MgSiO_3$) - ferrosilite ($FeSiO_3$), plus orthopyroxenes within sample 15386 ($MgSiO_3$ - $FeSiO_3$). [c]plagioclase = albite ($Na(AlSi_3O_8)$) - anorthite ($CaAl_2Si_2O_8$). [d]Olivine = Fayalite ($Fe_2SiO_4$) - Forsterite ($Mg_2SiO_4$). [e]ilmenite ($FeTiO_3$). [f]spinel = in general a solid solution between chromite ($Fe^{+2}Cr_2O_4$) and ulvöspinel ($Fe^{+2}{}_2TiO_4$) with a small proportion of hercynite ($Fe^{+2}Al_2O_4$). [g]armalcolite ($(Fe,Mg)Ti_2O_5$). [h]silica ($SiO_2$). [i]troilite (FeS). [j]mesostasis = interstisial material mostly too fine grained for analysis, frequently containing patches of K-feldspar ($KAlSi_3O_8$), glass and fine grained crystals of the preceding minerals. [k]Phosphates = apatite ($Ca_5(PO_4)_3(F,Cl)$ and (RE)-merillite ($Ca_{10}(Mg,Fe)_2(Y,REE)_2(PO_4)_{14}$). *Though the INCA phasemap tool has an error of ~ 1 % the modes of minor minerals were calculated based on phasemap data and SEM x-ray image pixel counting.

**Table 3:** Bulk rock major, minor (wt %), and trace (ppm) element chemistry of each sample, with 1σ errors.

| Mission | Apollo 11 | | | | | | | | | | | | | Apollo 12 | | | | | | | | | | | | | Apollo 14 | | Apollo 15 | | | | Apollo 17 | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Type | High-K | | | | | | | Low-K | | | | | | Olivine | | Ilmenite | | | | | | | | Pigeonite | | | High-Al | | Olivine Normative | | | | KREEP | | Type A | | Type B | | Type C | | Unclassified | | | |
| Sample | 10017_342 | 1σ | 10049_100 | 1σ | 10057_285 | 1σ | 10072_173 | 1σ | 10020_231 | 1σ | 10050_169 | 1σ | 10058_256 | 1σ | 12040_206 | 1σ | 12016_37 | 1σ | 12047_34 | 1σ | 12051_229 | 1σ | 12064_138 | 1σ | 12052_39 | 1σ | 14053_260 | 1σ | 15016_223 | 1σ | 15555_982 | 1σ | 15386_54 | 1σ | 75055_120 | 1σ | 70215_326 | 1σ | 74275_323 | 1σ | 70017_543 | 1σ | 70035_194 | 1σ |
| $SiO_2$ | 39.11 | 0.48 | 41.01 | 0.12 | 39.16 | 0.42 | 40.24 | 0.10 | 38.57 | 0.23 | 39.64 | 0.37 | 40.21 | 0.37 | 41.68 | 0.34 | 42.59 | 0.30 | 44.38 | 0.18 | 45.15 | 0.09 | 44.66 | 0.20 | 44.09 | 0.55 | 45.87 | 0.18 | 43.56 | 0.17 | 44.56 | 0.08 | 49.77 | 0.12 | 40.04 | 0.95 | 36.44 | 0.13 | 38.40 | 0.15 | 37.36 | 0.02 | 36.86 | 0.29 |
| $TiO_2$ | 11.72 | 0.05 | 10.82 | 0.17 | 11.27 | 0.07 | 11.89 | 0.16 | 10.69 | 0.11 | 11.68 | 0.17 | 8.66 | 0.05 | 2.66 | 0.03 | 3.72 | 0.01 | 4.98 | 0.06 | 4.45 | 0.04 | 3.66 | 0.04 | 3.29 | 0.04 | 2.60 | 0.02 | 2.09 | 0.01 | 1.10 | <0.01 | 2.05 | 0.01 | 12.91 | 0.08 | 13.32 | 0.06 | 12.74 | 0.07 | 13.38 | 0.17 | 13.73 | 0.17 |
| $Al_2O_3$ | 7.3 | 0.1 | 8.4 | <0.01 | 8.1 | 0.1 | 7.8 | <0.01 | 9.2 | 0.1 | 10.2 | 0.1 | 11.0 | 0.1 | 7.4 | 0.1 | 7.9 | 0.1 | 10.1 | 0.0 | 10.4 | <0.01 | 11.2 | 0.1 | 9.9 | <0.01 | 13.9 | 0.1 | 8.1 | <0.01 | 10.4 | <0.01 | 15.1 | 0.1 | 9.6 | 0.6 | 8.3 | <0.01 | 8.4 | <0.01 | 8.1 | <0.01 | 7.6 | 0.05 |
| FeO | 20.18 | 0.13 | 20.14 | 0.33 | 20.24 | 0.05 | 20.03 | 0.28 | 19.34 | 0.37 | 19.08 | 0.22 | 18.46 | 0.14 | 23.05 | 0.14 | 23.36 | 0.08 | 21.28 | 0.17 | 21.11 | 0.19 | 19.51 | 0.25 | 20.56 | 0.19 | 16.88 | 0.11 | 23.29 | 0.31 | 19.52 | 0.05 | 10.60 | 0.02 | 16.45 | 0.59 | 20.63 | 0.10 | 18.53 | 0.10 | 19.18 | 0.27 | 21.32 | 0.23 |
| MnO | 0.23 | <0.01 | 0.24 | <0.01 | 0.24 | <0.01 | 0.23 | <0.01 | 0.27 | <0.01 | 0.26 | <0.01 | 0.27 | <0.01 | 0.26 | <0.01 | 0.27 | <0.01 | 0.27 | <0.01 | 0.27 | <0.01 | 0.25 | <0.01 | 0.27 | <0.01 | 0.23 | <0.01 | 0.28 | <0.01 | 0.25 | <0.01 | 0.14 | <0.01 | 0.25 | <0.01 | 0.27 | <0.01 | 0.25 | <0.01 | 0.25 | <0.01 | 0.27 | <0.01 |
| MgO | 7.15 | 0.09 | 7.10 | 0.02 | 7.47 | 0.07 | 7.61 | 0.02 | 7.89 | 0.05 | 7.74 | 0.06 | 6.11 | 0.05 | 16.40 | 0.12 | 13.52 | 0.09 | 6.72 | 0.02 | 6.51 | 0.01 | 6.37 | 0.04 | 7.68 | 0.02 | 7.81 | 0.03 | 11.76 | 0.04 | 12.28 | 0.04 | 8.19 | 0.02 | 8.13 | 0.07 | 8.61 | 0.02 | 10.10 | 0.05 | 9.64 | 0.01 | 9.31 | 0.10 |
| CaO | 10.98 | 0.09 | 11.28 | 0.18 | 11.18 | 0.04 | 11.29 | 0.13 | 11.25 | 0.11 | 12.14 | 0.17 | 13.19 | 0.13 | 8.03 | 0.04 | 9.21 | 0.02 | 11.81 | 0.38 | 12.26 | 0.08 | 12.55 | 0.14 | 11.33 | 0.13 | 12.24 | 0.09 | 9.33 | 0.11 | 10.40 | 0.04 | 10.22 | 0.05 | 11.85 | 0.50 | 10.76 | 0.06 | 10.58 | 0.04 | 10.69 | 0.15 | 9.97 | 0.11 |
| $Na_2O$ | 0.48 | <0.01 | 0.56 | <0.01 | 0.54 | <0.01 | 0.51 | <0.01 | 0.39 | <0.01 | 0.40 | <0.01 | 0.46 | <0.01 | 0.21 | <0.01 | 0.24 | <0.01 | 0.32 | <0.01 | 0.32 | <0.01 | 0.31 | <0.01 | 0.27 | <0.01 | 0.49 | <0.01 | 0.25 | <0.01 | 0.30 | <0.01 | 0.90 | <0.01 | 0.43 | <0.01 | 0.39 | <0.01 | 0.42 | <0.01 | 0.38 | <0.01 | 0.39 | <0.01 |
| $K_2O$ | 0.32 | <0.01 | 0.34 | <0.01 | 0.32 | <0.01 | 0.31 | <0.01 | 0.06 | <0.01 | 0.07 | <0.01 | 0.09 | <0.01 | 0.04 | <0.01 | 0.05 | <0.01 | 0.07 | <0.01 | 0.07 | <0.01 | 0.06 | <0.01 | 0.07 | <0.01 | 0.12 | <0.01 | 0.04 | <0.01 | n.d | <0.01 | 0.64 | <0.01 | 0.05 | <0.01 | 0.04 | <0.01 | 0.08 | <0.01 | 0.04 | <0.01 | 0.08 | <0.01 |
| $P_2O_5$ | 0.16 | <0.01 | 0.18 | <0.01 | 0.17 | <0.01 | 0.16 | <0.01 | 0.08 | <0.01 | 0.05 | <0.01 | 0.08 | <0.01 | n.d | <0.01 | n.d | <0.01 | 0.07 | <0.01 | 0.08 | <0.01 | 0.06 | <0.01 | 0.07 | <0.01 | 0.14 | <0.01 | 0.05 | <0.01 | n.d | <0.01 | 0.72 | <0.01 | n.d | <0.01 | 0.08 | <0.01 | 0.05 | <0.01 | n.d | <0.01 | 0.06 | <0.01 |
| **Total** | **97.62** | | **100.07** | | **98.72** | | **100.05** | | **97.76** | | **101.31** | | **98.56** | | **99.78** | | **100.92** | | **99.99** | | **100.65** | | **98.65** | | **97.52** | | **100.29** | | **98.73** | | **98.85** | | **98.36** | | **99.71** | | **98.88** | | **99.56** | | **99.05** | | **99.56** | |
| **Mg#** | **38.71** | | **38.59** | | **39.67** | | **40.39** | | **42.10** | | **41.97** | | **37.10** | | **55.91** | | **50.78** | | **36.02** | | **35.46** | | **36.77** | | **39.97** | | **45.19** | | **47.36** | | **52.87** | | **57.94** | | **46.83** | | **42.65** | | **49.28** | | **47.26** | | **43.79** | |
| Li | 19.5 | 0.29 | 21.4 | 0.32 | 19.8 | 0.29 | 19.2 | 0.29 | 10.2 | 0.15 | 11.2 | 0.17 | 15.3 | 0.23 | 6.9 | 0.10 | 8.1 | 0.12 | 10.4 | 0.16 | 10.1 | 0.15 | 9.5 | 0.14 | 8.8 | 0.13 | 12.8 | 0.19 | 7.2 | 0.11 | 7.0 | 0.10 | 33.2 | 0.49 | 9.9 | 0.15 | 9.7 | 0.14 | 9.1 | 0.14 | 9.7 | 0.15 | 11.3 | 0.17 |
| Sc | 88.0 | 0.81 | 88.4 | 0.81 | 86.8 | 0.80 | 86.2 | 0.79 | 85.2 | 0.78 | 93.5 | 0.86 | 94.8 | 0.87 | 39.5 | 0.36 | 51.5 | 0.47 | 64.8 | 0.60 | 60.6 | 0.56 | 56.9 | 0.52 | 54.4 | 0.50 | 55.1 | 0.51 | 41.3 | 0.38 | 37.2 | 0.34 | 23.6 | 0.22 | 92.4 | 0.85 | 90.3 | 0.83 | 81.2 | 0.75 | 88.8 | 0.82 | 89.4 | 0.82 |
| V | 66.2 | 0.76 | 51.7 | 0.59 | 56.5 | 0.65 | 66.2 | 0.76 | 101.5 | 1.17 | 103.0 | 1.18 | 56.9 | 0.65 | 134.7 | 1.55 | 159.5 | 1.83 | 128.0 | 1.47 | 117.6 | 1.35 | 133.0 | 1.53 | 160.1 | 1.84 | 89.2 | 1.03 | 246.0 | 2.83 | 225.8 | 2.60 | 51.5 | 0.59 | 63.8 | 0.73 | 100.0 | 1.15 | 121.9 | 1.40 | 131.7 | 1.51 | 123.6 | 1.42 |
| Cr | 2334 | 28.01 | 2085 | 25.02 | 2187 | 26.24 | 2386 | 28.63 | 2614 | 31.37 | 2248 | 26.98 | 1410 | 16.92 | 3649 | 43.79 | 3683 | 44.20 | 1983 | 23.80 | 1722 | 20.66 | 1947 | 23.36 | 2992 | 35.90 | 2102 | 25.22 | 6075 | 72.90 | 4281 | 51.37 | 2040 | 24.48 | 2570 | 30.84 | 2677 | 32.12 | 3030 | 45.96 | 3698 | 44.38 | 3526 | 42.31 |
| Co | 27.4 | 0.39 | 26.4 | 0.37 | 26.9 | 0.38 | 27.8 | 0.39 | 18.4 | 0.26 | 15.0 | 0.21 | 11.4 | 0.16 | 65.0 | 0.92 | 57.0 | 0.80 | 34.2 | 0.48 | 29.8 | 0.42 | 25.1 | 0.35 | 36.2 | 0.51 | 24.0 | 0.34 | 60.4 | 0.85 | 57.0 | 0.80 | 19.8 | 0.28 | 15.5 | 0.22 | 22.7 | 0.32 | 21.4 | 0.30 | 23.9 | 0.34 | 20.4 | 0.29 |
| Ni | 5.7 | 0.06 | 5.9 | 0.06 | 5.2 | 0.05 | 7.6 | 0.08 | 3.6 | 0.04 | <2.6 | - | <2.6 | - | 83.5 | 0.89 | 58.5 | 0.62 | 12.0 | 0.13 | 7.5 | 0.08 | 3.3 | 0.04 | 14.1 | 0.15 | 10.9 | 0.12 | 89.5 | 0.95 | 83.1 | 0.88 | 9.2 | 0.10 | <2.6 | - | <2.6 | - | 4.4 | 0.05 | <2.6 | - | <2.6 | - |
| Ga | 5.8 | 0.07 | 6.5 | 0.08 | 6.0 | 0.07 | 5.8 | 0.07 | 3.9 | 0.05 | 4.4 | 0.05 | 5.3 | 0.06 | 2.7 | 0.03 | 3.3 | 0.04 | 4.3 | 0.05 | 4.2 | 0.05 | 4.0 | 0.05 | 3.6 | 0.04 | 4.0 | 0.05 | 3.4 | 0.04 | 3.1 | 0.04 | 7.7 | 0.09 | 3.4 | 0.04 | 3.7 | 0.04 | 3.6 | 0.04 | 3.7 | 0.04 | 4.5 | 0.05 |
| Rb | 6.1 | 0.14 | 7.0 | 0.16 | 6.3 | 0.14 | 6.1 | 0.14 | 0.8 | 0.02 | 0.7 | 0.02 | 0.9 | 0.02 | 0.7 | 0.02 | 0.7 | 0.02 | 1.0 | 0.02 | 1.1 | 0.02 | 1.2 | 0.03 | 1.3 | 0.03 | 2.4 | 0.05 | 0.7 | 0.02 | 0.3 | 0.01 | 17.6 | 0.39 | 0.4 | 0.01 | 0.4 | 0.01 | 1.2 | 0.03 | 0.4 | 0.01 | 0.8 | 0.02 |
| Sr | 173.9 | 3.01 | 191.5 | 3.31 | 177.3 | 3.07 | 167.7 | 2.90 | 144.4 | 2.50 | 177.3 | 3.07 | 207.8 | 3.59 | 90.7 | 1.57 | 119.1 | 2.06 | 157.8 | 2.73 | 153.9 | 2.66 | 135.9 | 2.35 | 115.3 | 1.99 | 107.2 | 1.85 | 87.8 | 1.52 | 85.4 | 1.48 | 186.2 | 3.22 | 149.8 | 2.59 | 121.4 | 2.10 | 159.7 | 2.76 | 160.0 | 2.77 | 194.6 | 3.37 |
| Y | 204.2 | 3.41 | 225.4 | 3.76 | 206.1 | 3.44 | 196.6 | 3.28 | 96.2 | 1.61 | 115.7 | 1.93 | 152.8 | 2.55 | 30.9 | 0.52 | 41.1 | 0.69 | 59.7 | 1.00 | 58.4 | 0.98 | 44.3 | 0.74 | 47.0 | 0.79 | 68.2 | 1.14 | 26.6 | 0.44 | 12.0 | 0.20 | 271.0 | 4.53 | 70.9 | 1.18 | 72.5 | 1.21 | 99.3 | 1.66 | 78.6 | 1.31 | 136.0 | 2.27 |
| Zr | 543.8 | 8.81 | 601.2 | 9.74 | 553.7 | 8.97 | 526.6 | 8.53 | 231.1 | 3.74 | 260.2 | 4.21 | 344.6 | 5.58 | 83.1 | 1.35 | 100.1 | 1.62 | 144.4 | 2.34 | 146.6 | 2.37 | 115.2 | 1.87 | 133.8 | 2.17 | 261.8 | 4.24 | 87.1 | 1.41 | 34.4 | 0.56 | 1247.0 | 20.20 | 174.5 | 2.83 | 192.1 | 3.11 | 254.8 | 4.13 | 212.8 | 3.45 | 350.1 | 5.67 |
| Nb | 31.9 | 0.62 | 35.1 | 0.68 | 32.3 | 0.62 | 23.6 | 0.45 | 19.0 | 0.37 | 21.7 | 0.42 | 26.9 | 0.52 | 5.5 | 0.11 | 5.8 | 0.11 | 8.2 | 0.16 | 8.8 | 0.17 | 7.7 | 0.15 | 9.3 | 0.18 | 19.4 | 0.37 | 6.1 | 0.12 | 2.3 | 0.05 | 82.9 | 1.60 | 18.3 | 0.35 | 21.9 | 0.42 | 23.6 | 0.45 | 21.1 | 0.41 | 32.5 | 0.63 |
| Cs | 0.17 | 0.01 | 0.19 | 0.01 | 0.17 | 0.01 | 0.16 | 0.01 | 0.03 | 0.00 | 0.02 | 0.00 | 0.03 | 0.00 | 0.03 | 0.00 | 0.03 | 0.00 | 0.04 | 0.00 | 0.05 | 0.00 | 0.05 | 0.00 | 0.05 | 0.00 | 0.10 | 0.00 | 0.03 | 0.00 | 0.01 | 0.00 | 0.73 | 0.03 | 0.01 | 0.00 | 0.01 | 0.00 | 0.05 | 0.00 | 0.01 | 0.00 | 0.03 | 0.00 |
| Ba | 288.80 | 3.96 | 329.60 | 4.52 | 300.20 | 4.11 | 278.50 | 3.82 | 70.94 | 0.97 | 76.16 | 1.04 | 102.30 | 1.40 | 45.23 | 0.62 | 44.95 | 0.62 | 66.89 | 0.92 | 72.18 | 0.99 | 65.96 | 0.90 | 75.82 | 1.04 | 177.90 | 2.44 | 47.88 | 0.66 | 20.08 | 0.28 | 823.80 | 11.29 | 48.10 | 0.66 | 58.20 | 0.80 | 69.20 | 0.95 | 55.58 | 0.76 | 101.70 | 1.39 |
| La | 25.83 | 0.39 | 29.10 | 0.44 | 26.56 | 0.40 | 24.93 | 0.38 | 7.63 | 0.12 | 7.48 | 0.11 | 10.20 | 0.16 | 4.04 | 0.06 | 4.16 | 0.06 | 6.66 | 0.10 | 6.90 | 0.10 | 5.86 | 0.09 | 7.03 | 0.11 | 14.64 | 0.22 | 4.77 | 0.07 | 1.79 | 0.03 | 78.33 | 1.19 | 3.66 | 0.06 | 4.94 | 0.08 | 6.10 | 0.09 | 4.22 | 0.06 | 7.97 | 0.12 |
| Ce | 76.78 | 1.01 | 86.13 | 1.13 | 79.38 | 1.04 | 74.58 | 0.98 | 24.05 | 0.32 | 26.35 | 0.35 | 35.39 | 0.46 | 11.49 | 0.15 | 12.49 | 0.16 | 19.34 | 0.25 | 20.09 | 0.26 | 16.94 | 0.22 | 19.56 | 0.26 | 38.25 | 0.50 | 13.15 | 0.17 | 4.94 | 0.06 | 200.40 | 2.63 | 12.84 | 0.17 | 15.98 | 0.21 | 21.28 | 0.28 | 14.85 | 0.19 | 27.83 | 0.36 |
| Pr | 11.71 | 0.15 | 13.09 | 0.17 | 11.99 | 0.15 | 11.27 | 0.14 | 4.14 | 0.05 | 4.94 | 0.06 | 6.61 | 0.08 | 1.78 | 0.02 | 2.04 | 0.03 | 3.18 | 0.04 | 3.24 | 0.04 | 2.60 | 0.03 | 2.98 | 0.04 | 5.43 | 0.07 | 2.01 | 0.03 | 0.76 | 0.01 | 27.27 | 0.35 | 2.43 | 0.03 | 2.83 | 0.04 | 3.97 | 0.05 | 2.81 | 0.04 | 5.24 | 0.07 |
| Nd | 58.16 | 0.70 | 65.07 | 0.79 | 59.74 | 0.72 | 56.22 | 0.68 | 22.80 | 0.28 | 28.33 | 0.34 | 37.76 | 0.46 | 8.90 | 0.11 | 10.63 | 0.13 | 16.26 | 0.20 | 16.46 | 0.20 | 12.97 | 0.16 | 14.51 | 0.18 | 24.37 | 0.29 | 9.79 | 0.12 | 3.82 | 0.05 | 119.75 | 1.45 | 14.44 | 0.17 | 15.71 | 0.19 | 22.86 | 0.28 | 16.48 | 0.20 | 30.11 | 0.36 |
| Sm | 20.31 | 0.37 | 22.74 | 0.41 | 20.93 | 0.38 | 19.68 | 0.35 | 9.00 | 0.16 | 11.70 | 0.21 | 15.56 | 0.28 | 3.18 | 0.06 | 4.01 | 0.07 | 6.04 | 0.11 | 6.08 | 0.11 | 4.64 | 0.08 | 5.05 | 0.09 | 7.47 | 0.13 | 3.27 | 0.06 | 1.34 | 0.02 | 34.10 | 0.61 | 6.35 | 0.11 | 6.54 | 0.12 | 9.59 | 0.17 | 7.16 | 0.13 | 12.64 | 0.23 |
| Eu | 2.17 | 0.04 | 2.39 | 0.05 | 2.19 | 0.04 | 2.10 | 0.04 | 1.55 | 0.03 | 2.03 | 0.04 | 2.49 | 0.05 | 0.78 | 0.02 | 0.99 | 0.02 | 1.39 | 0.03 | 1.35 | 0.03 | 1.13 | 0.02 | 1.05 | 0.02 | 1.31 | 0.03 | 0.80 | 0.02 | 0.61 | 0.01 | 2.66 | 0.05 | 1.50 | 0.03 | 1.34 | 0.03 | 1.78 | 0.03 | 1.67 | 0.03 | 2.31 | 0.05 |
| Gd | 26.49 | 0.35 | 29.37 | 0.39 | 27.16 | 0.36 | 25.65 | 0.34 | 12.39 | 0.16 | 15.85 | 0.21 | 20.88 | 0.28 | 4.18 | 0.06 | 5.45 | 0.07 | 8.07 | 0.11 | 8.03 | 0.11 | 6.05 | 0.08 | 6.51 | 0.09 | 9.06 | 0.12 | 4.17 | 0.06 | 1.76 | 0.02 | 39.03 | 0.52 | 9.17 | 0.12 | 9.21 | 0.12 | 13.13 | 0.17 | 10.14 | 0.13 | 17.50 | 0.23 |
| Tb | 4.96 | 0.12 | 5.57 | 0.13 | 5.09 | 0.12 | 4.83 | 0.11 | 2.36 | 0.06 | 2.99 | 0.07 | 3.98 | 0.09 | 0.79 | 0.02 | 1.05 | 0.02 | 1.54 | 0.04 | 1.52 | 0.04 | 1.15 | 0.03 | 1.23 | 0.03 | 1.71 | 0.04 | 0.75 | 0.02 | 0.33 | 0.01 | 6.97 | 0.16 | 1.81 | 0.04 | 1.79 | 0.04 | 2.52 | 0.06 | 1.98 | 0.05 | 3.38 | 0.08 |
| Dy | 32.02 | 0.55 | 35.47 | 0.61 | 32.79 | 0.57 | 31.10 | 0.54 | 15.43 | 0.27 | 19.14 | 0.33 | 25.39 | 0.44 | 5.15 | 0.09 | 6.80 | 0.12 | 9.99 | 0.17 | 9.84 | 0.17 | 7.36 | 0.13 | 7.87 | 0.14 | 10.85 | 0.19 | 4.60 | 0.08 | 2.04 | 0.04 | 43.26 | 0.75 | 11.81 | 0.20 | 11.84 | 0.20 | 16.16 | 0.28 | 13.00 | 0.22 | 22.00 | 0.38 |
| Ho | 7.02 | 0.17 | 7.72 | 0.19 | 7.11 | 0.17 | 6.78 | 0.16 | 3.37 | 0.08 | 4.10 | 0.10 | 5.47 | 0.13 | 1.11 | 0.03 | 1.49 | 0.04 | 2.18 | 0.05 | 2.13 | 0.05 | 1.60 | 0.04 | 1.70 | 0.04 | 2.41 | 0.06 | 0.96 | 0.02 | 0.43 | 0.01 | 9.35 | 0.23 | 2.60 | 0.06 | 2.63 | 0.06 | 3.50 | 0.08 | 2.84 | 0.07 | 4.76 | 0.12 |
| Er | 19.49 | 0.33 | 21.62 | 0.37 | 19.93 | 0.34 | 18.91 | 0.32 | 9.48 | 0.16 | 11.23 | 0.19 | 14.92 | 0.26 | 3.13 | 0.05 | 4.14 | 0.07 | 6.04 | 0.10 | 5.90 | 0.10 | 4.43 | 0.08 | 4.77 | 0.08 | 6.92 | 0.12 | 2.56 | 0.04 | 1.18 | 0.02 | 26.16 | 0.45 | 7.31 | 0.13 | 7.41 | 0.13 | 9.67 | 0.17 | 8.03 | 0.14 | 13.18 | 0.23 |
| Yb | 17.46 | 0.29 | 19.21 | 0.32 | 17.80 | 0.29 | 16.87 | 0.28 | 8.65 | 0.14 | 9.92 | 0.16 | 13.11 | 0.22 | 2.80 | 0.05 | 3.73 | 0.06 | 5.41 | 0.09 | 5.22 | 0.09 | 3.94 | 0.06 | 4.23 | 0.07 | 6.47 | 0.11 | 2.13 | 0.04 | 1.00 | 0.02 | 23.04 | 0.38 | 6.72 | 0.11 | 6.89 | 0.11 | 8.60 | 0.14 | 7.42 | 0.12 | 11.98 | 0.20 |
| Lu | 2.53 | 0.04 | 2.80 | 0.04 | 2.61 | 0.04 | 2.46 | 0.03 | 1.30 | 0.02 | 1.46 | 0.02 | 1.91 | 0.03 | 0.41 | 0.01 | 0.55 | 0.01 | 0.79 | 0.01 | 0.77 | 0.01 | 0.58 | 0.01 | 0.63 | 0.01 | 0.96 | 0.01 | 0.31 | 0.00 | <0.16 | - | 3.35 | 0.05 | 1.02 | 0.01 | 1.05 | 0.01 | 1.27 | 0.02 | 1.11 | 0.02 | 1.75 | 0.02 |
| Hf | 15.97 | 0.23 | 17.69 | 0.25 | 16.48 | 0.24 | 15.53 | 0.22 | 7.34 | 0.11 | 8.47 | 0.12 | 11.09 | 0.16 | 2.42 | 0.03 | 3.12 | 0.04 | 4.44 | 0.06 | 4.38 | 0.06 | 3.42 | 0.05 | 3.77 | 0.05 | 6.15 | 0.09 | 2.35 | 0.03 | 0.97 | 0.01 | 27.43 | 0.40 | 5.90 | 0.09 | 6.34 | 0.09 | 8.46 | 0.12 | 7.01 | 0.10 | 11.26 | 0.16 |
| Pb | 1.78 | 0.03 | 2.27 | 0.03 | 2.03 | 0.03 | 1.86 | 0.03 | <0.99 | - | <0.99 | - | <0.99 | - | <0.99 | - | <0.99 | - | <0.99 | - | <0.99 | - | <0.99 | - | <0.99 | - | 2.35 | 0.04 | <0.99 | - | <0.99 | - | 8.84 | 0.13 | <0.99 | - | <0.99 | - | <0.99 | - | <0.99 | - | <0.99 | - |
| Ta | 1.99 | 0.03 | 2.13 | 0.04 | 1.89 | 0.03 | 1.41 | 0.02 | 1.28 | 0.02 | 1.18 | 0.02 | 1.71 | 0.03 | 0.32 | 0.01 | 0.34 | 0.01 | 0.48 | 0.01 | 0.52 | 0.01 | 0.44 | 0.01 | 0.51 | 0.01 | 1.00 | 0.02 | 0.38 | 0.01 | 0.16 | 0.00 | 3.45 | 0.06 | 1.32 | 0.02 | 1.55 | 0.03 | 1.66 | 0.03 | 1.46 | 0.03 | 2.17 | 0.04 |
| Th | 3.63 | 0.10 | 4.11 | 0.12 | 3.73 | 0.11 | 3.49 | 0.10 | 0.72 | 0.02 | 0.56 | 0.02 | 0.84 | 0.02 | 0.60 | 0.02 | 0.56 | 0.02 | 0.82 | 0.02 | 0.95 | 0.03 | 0.84 | 0.02 | 1.09 | 0.03 | 2.39 | 0.07 | 0.52 | 0.01 | 0.18 | 0.01 | 14.33 | 0.41 | 0.25 | 0.01 | 0.40 | 0.01 | 0.50 | 0.01 | 0.28 | 0.01 | 0.52 | 0.01 |
| U | 0.88 | 0.01 | 1.00 | 0.01 | 0.92 | 0.01 | 0.84 | 0.01 | 0.19 | 0.00 | 0.15 | 0.00 | 0.24 | 0.00 | 0.16 | 0.00 | 0.15 | 0.00 | 0.22 | 0.00 | 0.25 | 0.00 | 0.22 | 0.00 | 0.29 | 0.00 | 0.62 | 0.01 | 0.14 | 0.00 | <0.09 | - | 3.72 | 0.05 | <0.09 | - | 0.11 | 0.00 | 0.14 | 0.00 | <0.09 | - | 0.15 | 0.00 |

Trace element errors were calculated based on 1σ values for five repeat analyses of the standard monitor material DNC-1. The 1σ errors of each trace element analysis were calculated by converting DNC-1 1σ values to % difference values, and then calculating the ppm equivalent for each individual elemental analysis (see also SOM, Table S2). Where < precedes a value the value shown is the detection limit, and the abundance of that particular element is below the detection limit in that sample.

**Table 4**: Estimated parental melt REE compositions for mare basalts analysed in this study.

| | Apollo 12 | | | Apollo 14 | Apollo 15 | Apollo 11 | | Apollo 17 | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Pig Parent | Olv parent | Ilm parent | Group C | Olv Norm | Type B3 | Orange Glass[g] | Type A | Type B2 | Type C |
| REE | 12011[a] | Average[b] | Average[c] | Hybrid[d] | 15537[e] | 10045[f] | | Estimated[h] | Estimated[h] | Average[i] |
| La | 33.11 | 24.76 | 26.91 | 26.21 | 22.18 | 28.55 | 26.42 | 20.45 | 18.75 | 26.84 |
| Ce | 30.67 | 27.19 | 28.11 | 20.91 | 24.20 | 37.30 | 31.50 | | | 36.14 |
| Pr | 31.31 | 28.90 | 32.92 | | | | | | | |
| Nd | 31.39 | 31.83 | 35.32 | | | 46.64 | 39.35 | | | 52.61 |
| Sm | 32.63 | 29.50 | 39.26 | 10.97 | 24.58 | 57.10 | 44.19 | 47.59 | 39.43 | 64.58 |
| Eu | 19.82 | 17.50 | 21.93 | 6.46 | 14.29 | 26.79 | 32.14 | 27.68 | 21.43 | 32.14 |
| Gd | 26.55 | 26.07 | 35.97 | | | 67.14 | | | | 75.28 |
| Tb | 25.90 | 27.55 | 38.55 | | 21.49 | 57.85 | 45.45 | | | |
| Dy | 24.19 | 26.12 | 36.82 | | | 59.74 | 38.73 | | | 69.63 |
| Ho | 23.74 | 23.74 | 32.91 | | | | | | | |
| Er | 23.47 | 23.54 | 32.49 | | | 61.04 | | | | 61.04 |
| Tm | 21.07 | 21.69 | 29.81 | | | | | | | |
| Yb | 23.75 | 20.43 | 29.31 | 9.91 | 14.00 | 52.31 | 27.26 | 43.69 | 36.31 | 54.15 |
| Lu | 19.34 | 19.55 | 26.99 | 8.89 | 12.80 | 49.38 | 25.10 | | | 49.38 |

[a]Snyder et al. (1997). [b]Average of samples 12009 and 12015 (Snyder et al. 1997). [c]Average of samples 12008, 12022 and 12045 (Snyder et al. 1997). [d]Hybrid taking the lowest elemental abundances for all samples within this group (Neal and Kramer, 2006). [e]Vetter et al. (1989). [f]Agrell et al. (1970). [g]Jerde et al. (1994). [h]Estimation based on element-element plots (Neal et al., 1990b). [i]Average based on samples 74245, 74275 and 74247 (Snyder et al., 1992). All data are chondrite normalised based on the data of Anders and Grevesse (1989).

**Table 5**: REE abundances (in ppm) in the mare basalt source regions and assimilants used for chemical modelling

| Element | Low-Ti source no TIRL[1] | Low-Ti source 1% TIRL[1] | Low-Ti source 2% TIRL[1] | High-Ti source[2] | High-Al source[3] | High-K KREEP[4] | A14 KREEP/granitic assimilant[5] | A14 granitic assimilant[6] |
|---|---|---|---|---|---|---|---|---|
| La | 0.004 | 0.08 | 0.42 | 0.27 | 0.15 | 110 | 119 | 58 |
| Ce | 0.02 | 0.22 | 1.09 | 0.80 | 0.37 | 280 | 288 | 149 |
| Pr | 0.005 | 0.03 | 0.16 | 0.15 | 0.05 | 37 | | |
| Nd | 0.04 | 0.19 | 0.81 | 0.90 | 0.28 | 178 | | |
| Sm | 0.03 | 0.07 | 0.27 | 0.39 | 0.10 | 48 | 49 | 22 |
| Eu | 0.01 | 0.02 | 0.05 | 0.07 | 0.03 | 3 | 2 | 3.3 |
| Gd | 0.07 | 0.12 | 0.37 | 0.58 | 0.15 | 58 | | |
| Tb | 0.02 | 0.03 | 0.07 | 0.11 | 0.03 | 10 | | |
| Dy | 0.13 | 0.20 | 0.49 | 0.81 | 0.21 | 65 | | |
| Ho | 0.04 | 0.05 | 0.12 | 0.20 | 0.05 | 14 | | |
| Er | 0.14 | 0.18 | 0.35 | 0.60 | 0.15 | 40 | | |
| Tm | 0.03 | 0.03 | 0.06 | 0.09 | 0.02 | 6 | | |
| Yb | 0.21 | 0.24 | 0.39 | 0.64 | 0.17 | 36 | 48 | 18 |
| Lu | 0.04 | 0.04 | 0.06 | 0.10 | 0.03 | 5 | 7 | 2.6 |

[1]Data digitised from Snyder et al. (1992a), Fig. 6. [2]Data digitised from Snyder et al. (1994), Fig. 11. Data digitised from Hughes et al. (1990), Fig. 1. [4]Waren (1989). [5]Neal and Kramer (2006) granitic assimilant, consisting of a mixture of the REE compositions of Apollo 14 granite samples 14030,204 and 14321,1027 (Warren et al., 1983), and quartz monzodiorite sample 14161,7069 (Jolliff, 1991), in the proportions 1:5:4. [6]Granite 14303,204 (Warren et al., 1983).

**Table 6:** Mineral REE partition coefficients used in this study.

| | Partition Coefficients | | | | | | |
|---|---|---|---|---|---|---|---|
| | **olivine**[1] | **orthopyroxene**[2] | **augite**[2] | **pigeonite**[3] | **plagioclase**[4] | **ilmenite**[5] | **garnet**[8] |
| **La** | 0.0001 | 0.0007 | 0.0446 | 0.0009 | 0.0418 | 0.001 | 0.0014 |
| **Ce** | 0.0001 | 0.0015 | 0.0733 | 0.00172 | 0.0302 | 0.0062 | 0.0029 |
| **Pr** | | | | | | | |
| **Nd** | 0.0001 | 0.0055 | 0.1544 | 0.0058 | 0.0236 | 0.00077 | 0.03 |
| **Sm** | 0.0006 | 0.0143 | 0.251 | 0.011 | 0.017 | 0.0026 | 0.18 |
| **Eu** | 0.0007 | 0.0204 | 0.2952 | 0.0068 | 0.166 | 0.0015 | 0.33 |
| **Gd** | 0.001 | 0.0281 | 0.3377 | 0.021 | 0.0105 | 0.013[6] | 0.75 |
| **Tb** | 0.002 | 0.0376 | 0.3758 | 0.027 | 0.0095 | 0.0082[7] | 1.5 |
| **Dy** | 0.003 | 0.0487 | 0.4071 | 0.034 | 0.0089 | 0.013[7] | 2.4 |
| **Ho** | | | | | | | |
| **Er** | 0.008 | 0.0714 | 0.4402 | 0.055 | 0.0077 | 0.019 | 4.4 |
| **Tm** | | | | | | | |
| **Yb** | 0.019 | 0.0913 | 0.4426 | 0.087 | 0.0065 | 0.0045 | 6.5 |
| **Lu** | 0.03 | 0.0995 | 0.4368 | 0.11 | 0.0068 | 0.058 | 6.7 |

[1]McKay et al. (1986). [2]Yao et al. 2012. [3]McKay et al. (1991). [4]Phinney and Morrison (1990). [5]van Kan Parker (2011) unless otherwise indicated. [6]Klemme et al. (2006). [7]Nakamura et al. (1986). [8]Green et al. (2000).

**Table 7:** Chemical modelling parameters that produce the best match between measured and modelled parental melt REE abundances.

| | Type | Sample | Source | Mineral Modes in Source | Partial Melt | Crystal Fractionation/Assimilation |
|---|---|---|---|---|---|---|
| Apollo 11 | High-K | 10017<br>10049<br>10057<br>10072 | 80:20 mix of 86 and 95 PCS + 2% TIRL | 49% olv, 23% opx, 23% pig, 2% aug, 3% ilm | 5% fractional | 5% A/FC (r = 0.8): KREEP/(50 % ilm, 30 % pig, 20 % aug) |
| | Low-K | 10020<br>10050<br>10058 | 80:20 mix of 86 and 95 PCS + 1% TIRL | 49% olv, 23% opx, 23% pig, 2% aug, 3% ilm | 3-5% fractional | |
| Apollo 12 | Olivine | 12040 | 86 PCS + <1% TIRL | 52% olv, 23% opx, 23% pig, 2% aug | 1-3% fractional | |
| | Ilmenite | 12016<br>12047<br>12051<br>12064 | | 53% olv, 23% opx, 23% pig, 1% plag | 1-2% fractional | |
| | Pigeonite | 12052 | | 52% olv, 23% opx, 23% pig, 2% aug | 1-3% fractional | |
| Apollo 14 | High-Al | 14053 | Hughes et al. (1990) PCS with 0.76% TIRL and 3% plag | 49% olv, 23% opx, 23% pig, 2% aug, 3% plag | 1% batch* | 40 % A/FC (r = 0.1): granitic asimilant/100 % olv* |
| Apollo 15 | Olv. Norm. | 15016<br>15555 | 86 PCS + 2% TIRL | 52% olv, 23% opx, 23% pig, 2% aug, ±0.5% gar | 3-9% batch | |
| Apollo 17 | Type A | 75055 | <95 PCS component and/or < 1% TIRL | 49% olv, 23% opx, 23% pig, 2% aug, 3% ilm | 4-6% fractional | |
| | Type B | 70215 | 80:20 mix of 86 and 95 PCS + 1% TIRL | | 3-6% batch | |
| | Type C | 74275 | | | 2-4% fractional | |

**Fig. 1:** Bulk-rock $TiO_2$ (wt%) vs. **(a)** $Al_2O_3$ (wt%), **(b)** $K_2O$ (wt%) and **(c)** MgO (wt%). Symbols represent our data, envelopes represent previously published data for Apollo mare basalts. 1σ errors are generally smaller than the symbol size. Source: **Apollo 11** – LSPET, 1969; Agrell et al., 1970; Compston et al., 1970; Engel and Engel, 1970; Goles et al., 1970a; Goles et al., 1970b; Maxwell et al., 1970; Morrison et al., 1970; Turekian and Kharkar, 1970; Wakita et al., 1970; Wanke et al., 1970; Engel et al., 1971; Wanke et al., 1971; Kharkar and Turekian, 1971; Duncan et al., 1976; Beaty and Albee, 1978; Beaty et al., 1979a; Ma et al., 1980; Rhodes and Blanchard, 1980; Jerde et al., 1994; Neal et al., 1994a. **Apollo 12** – LSPET, 1970; Maxwell et al., 1970; Compston et al., 1971; Engel et al., 1971; Goles et al., 1971; Klein et al., 1971; Scoon, 1971; Smales et al., 1971; Warner, 1971; Baldridge et al., 1979; Beaty et al., 1979b; Warren et al., 1986; Neal et al., 1994a; Snyder et al., 1997. **Apollo 14** – LSPET, 1971; Ehmann et al., 1972; Hubbard et al., 1972; Laul et al., 1972; Philpotts et al., 1972; Rose et al., 1972; Taylor et al., 1972; Wanke et al., 1972; Willis et al., 1972; Helmke et al., 1973; Duncan et al., 1975; Vaniman and Papike, 1980; Warner et al., 1980; Taylor et al., 1983; Dickinson et al., 1985; Shervais et al., 1985a; Warren et al., 1986; Neal et al., 1988a, b; Neal et al., 1989. **Apollo 15** – Chappell and Green, 1973; Rhodes and Hubbard, 1973; Ma et al., 1978; Ryder and Steele, 1988; Vetter et al., 1989. **Apollo 15 KREEP** - Rhodes and Hubbard, 1973; Basu and Bower, 1976; Irving, 1977; Lindstrom et al., 1977; Warren et al., 1983; Simon et al., 1988; Ryder and Martinez, 1991. **Apollo 17** – Brunfelt et al., 1974; Duncan et al., 1974; Rhodes et al., 1974; Rose et al., 1974; Shih et al., 1975; Wanke et al., 1975; Warner et al., 1975; Rhodes et al., 1976; Murali et al., 1977; Warner et al., 1979; Neal et al., 1990a,b; Ryder, 1990. **Lunar basaltic meteorites** – Warren and Kellemeyn, 1993; Anand et al., 2003, 2006; Day et al., 2006; Haloda et al., 2006; Joy et al., 2006; Joy et al., 2008.

**Fig. 2:** Compatible trace element plots including previously published data. Mg# vs. Sc (ppm) **(a)**, Mg# vs. V (ppm) **(b)** and Mg# vs. Cr (ppm) **(c)**. Large symbols represent data from this study; small symbols represent previously published data. 1σ errors are smaller than the symbol sizes for all data points. The negative Sc vs. Mg# trend for Apollo 12 mare basalts is highlighted by a grey envelope **(a)**. Positive correlations between V and Mg# for the Apollo 11 and 17 mare basalts are highlighted by light grey and dark grey envelopes, respectively **(b)**. Similarly, positive correlations between Cr and Mg# for the Apollo 12 and 17 mare basalts are highlighted by light grey and dark grey envelopes, respectively **(c)**. Lunar basaltic meteorites are represented by the black outline. Previously published data references are the same as for Fig. 1.

**Fig. 3:** Chondrite-normalised bulk-rock REE patterns for all samples. Apollo 15 sample 15555,982 data (labelled) is disregarded as being unrepresentative. 1σ errors are smaller than the symbol size.

**Fig. 4:** Incompatible trace element bi-variate ratio plots (ppm). Apollo 15 KREEP basalt 15386 consistently displays the highest incompatible element abundance, followed by the Apollo 11 high-K basalts. Apollo 14 high-Al basalt 14053 also contains elevated K, Rb, Cs, Ba, La, Th and U contents compared to the other mare basalts. These plots highlight compositional variations between the high- and low-Ti mare basalts, as well as between the individual Apollo 11 and Apollo 17 basalt groups. Excluding sample 15555,982, the Apollo 12 and 15 low-Ti basalts in our sample set show similar incompatible element abundances, with increasing abundances within the more evolved samples. The uncharacteristically low incompatible element abundances in sample 15555,982 suggest this sample has a

heterogeneous composition (as also reported by Ryder and Schuraytz, 2001 and Spicuzza et al., 2007). 1σ errors are smaller than the symbol size for all data points.

**Fig. 5:** Chondrite-normalised REE pattern plots comparing mare basalt parental melt (Table 4) and measured sample compositions. **(a)** Apollo 12 and 15 low-Ti basalts, **(b)** Apollo 14 high-Ti basalts and **(c)** Apollo 11 and 17 high-Ti basalts. Parental melt compositions are represented by dashed lines, sample compositions are represented by solid lines. The chondritic REE values of Anders and Grevesse (1989) were used for normalisation.

**Fig. 6:** Chondrite-normalised REE pattern plots showing our calculated source compositions and partial melts, compared to the mare basalt parental melt compositions (Table 4). Apollo 12 olivine and pigeonite basalts (**a**) and ilmenite basalt (**b**) partial melts were calculated based on a two sources of 86 PCS with 1% TIRL and 86 PCS with no TIRL. Apollo 15 olivine normative partial melts were calculated for source compositions with 1 % and 2 % TIRL (**c**), plus with 0.5 % garnet in the source **(d**). The Apollo 11 low-K (**e**) and Apollo 17 basalt partial melts were calculated based on source regions with a 80:20 mix of 86 and 95 PCS, with 1% TIRL **(f-h**). The chondritic REE values of Anders and Grevesse (1989) were used for normalisation. Note the different scales on the y-axis.

**Fig. 7:** Chondrite-normalised REE pattern plots, showing assimilation/fractional crystallisation (AFC) calculation results. The chondritic REE values of Anders and Grevesse (1989) were used for normalisation. Note the different scales on the y-axis. Orange glass data from Jerde et al. (1994) (Table 4).